Final Report
An Evaluation of Feasibility of Light Water Reactor (LWR) Based Actinide Transmutation Concepts

Transmutation of Actinides in PWRs Using Fertile Free Fuels

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Abstract

A preliminary assessment of the neutronics of PWR cores containing fertile free fuel (FFF) hosting Trans-Uranic Nuclides (TRU) from the spent fuel of current Light Water Reactors has been conducted. First, the ability to burn down the TRUs in homogeneous and heterogeneous cores is assessed. The results indicate that under the constraints of current power density, cycle length, and safety requirements the once-through TRU burner based on FFF can reduce the initial amount of TRU by approximately a factor of two. However, the effect of the once-through TRU burner on radiotoxicity of the fuel going to the repository was found to be limited. The Doppler coefficient of these fuels is negative throughout the cycle, albeit it exhibits smaller values than those of typical UO_2 and MOX fuels. Moderator temperature and coolant void worth coefficients are negative and comparable in values to UO_2 fuel. Boron worth is significantly lower than for UO_2 fuel due to a harder neutron spectrum, but comparable to MOX fuel. At the same time, the BOL reactivity excess is significantly smaller than that for UO_2 fuels requiring less reactivity compensation. A considerably lower effective delayed neutron fraction than that of typical UO_2 and MOX fuel is the key challenge for control of a PWR core fully loaded with FFF assemblies.

Secondly, the feasibility of a conventional PWR operating in a sustainable fuel cycle mode with complete recycling of TRU elements in the same reactor was investigated. A new Combined Non-fertile and Uranium (CONFU) assembly where some of the uranium fuel pins are replaced with FFF pins destined for destruction of TRU generated in the previous cycle has been proposed and analyzed. In contrast to the burn down scenario, the sustainable fuel cycle option can, for the same amount of energy generation, significantly reduce both the amounts and radiotoxicity of the nuclear waste in comparison with the conventional once-through UO_2 fuel cycle. It is shown that under the constraints of acceptable power peaking limits, the CONFU assembly exhibits negative reactivity feedback coefficients comparable in values to those of the reference UO2 fuel. Moreover, the effective delayed neutron fraction is about the same as for UO2-fueled cores. Therefore, feasibility of the PWR core design with complete TRU recycle has been shown in principle. However, gradual build up of small amounts of Cm and Cf challenges fuel cycle reprocessing and fabrication stages due to the high spontaneous fissions rates of these nuclides. Feasibility of the processing steps and the implications for the entire fuel cycle will have to be addressed in the future.

Acknowledgement

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Table of Contents

List of Figures

List of Tables

Neutronic Assessment of Fertile Free Fuels

1. Introduction

The destruction of Pu and MA in LWRs using mixed oxides with UO_2 are being studied in many parts of the world. The transmutation rates can be effectively accelerated provided that the reactor core contains limited amounts of a fertile U-238. Therefore, the fuels that contain inert matrix material as a substitute for fertile (UO_2) matrix can perform the task of Trans-Uranic Elements (TRU) destruction in the most effective way.

Numerous studies carried out in the past have shown that the Reactors Grade (RG) and Weapons Grade (WG) Pu can be effectively burned in the fertile-free fuels (FFF) while maintaining comparable to current generation PWRs the reactivity control and safety characteristics of the fuel. ([1],[2],[3],[4] and others)

Pu or TRU can be either mixed homogeneously with or dispersed as micro-size particles in the inert matrix. The dispersed micro-particles approach provides additional flexibility in the choice of matrix materials. The matrix and the micro-particle materials can be separately chosen so that in combination they will provide good mechanical and chemical stability, radiation damage resistance, compatibility with the cladding material and water coolant in addition to good thermal properties and low parasitic neutron absorption. The matrix material has also to be chemically stable in the nuclear waste repository environment, at the same time it should preferably allow a simple and inexpensive reprocessing.

The focus of this part of the work is mainly on a preliminary neutronic evaluation of the TRU containing FFFs. In particular, $MgAl_2O_4$ (Spinel) was chosen as a primary host matrix material and Yttria Stabilized Zirconia (YSZ) was chosen to be a part of the micro-spheres composition in order to enhance the irradiation and mechanical stability of the fuel particles. The detailed arguments promoting the choice of these inert materials to be used in the current study are described in Appendix A and Reference [5].

The reported work includes several parts. First, a series of benchmark calculations were performed using available computational tools in order to evaluate their capability to handle non-conventional fuel designs with large loadings of TRUs. In the evaluation of FFF, two conceptually different fuel cycle scenarios were considered: a TRU burn-down scenario aiming at the effective and fast reduction of the existing TRU stockpile and a sustainable fuel cycle scenario that would have a complete recycling of Pu and MA, that is, zero net generation of TRU and, therefore, negligible impact on the environment. The following fuel cycle parameters were calculated for each scenario: destruction rates and residual fractions of Pu and MA in the spent fuel, spent fuel radiotoxicity, decay heat load and a neutron source from spontaneous fissions and (α,n) reactions. In addition, reactivity feedback coefficients were calculated for each of the options analyzed to assess the feasibility of using FFFs in the current generation of PWRs.

1.1 Methodology and Description of Calculated Cases.

All the burnup and criticality calculations in this study were performed using the CASMO4 fuel assembly burnup and 2D transport computer code [6], which uses a 70 energy group neutron cross-section library. The CASMO4 code tracks the evolution of about 200 fission products and over 40 actinides ranging from Th-232 to Cf-252.

The reactivity limited single batch burnup (BU1) and fuel cycle length were estimated by calculating the burnup at which k_{inf} of the fuel equals to 1.03 allowing 3% reactivity penalty for the core leakage. This value is typical for typical PWR cores employing the low-leakage fuel loading pattern. However, the TRU containing cores are expected to have higher leakage due to the harder spectrum than for a typical PWR neutron spectrum. Therefore, the calculated BU1 values are somewhat of an overestimate. The discharge fuel burnup was calculated as 1.5×BU1 assuming a 3-batch core fuel management and linear dependence of the batch reactivity on the burnup [7].

All burnup calculations were performed for the fuel assembly with reference geometry and reference PWR operating conditions (Table 1). The initial TRU isotopic vector used in all calculated cases is shown in Table 2.

Table 1. Reference Fuel Assembly Geometry and Operating Conditions

Number of Fuel Locations per Assembly	264
Number of Guide Tube Locations	25
Fuel Assembly Pitch, cm	21.5
Fuel Assembly Gap, cm	0.08
Fuel pellet radius, cm	0.4095
Gap thickness, cm	0.0083
Cladding Outer Radius, cm	0.4750
Lattice Pitch, cm	1.26
Active Fuel Height, cm	366
Guide Tube Inner Radius, cm	0.5715
Guide Tube Outer Radius, cm	0.6120
Cladding Material	Zircaloy-4
Cladding Density, g/cm^3	6.55
Fuel temperature, K	900
Coolant temperature, K	580
Core Average Power density, kW/l	104
System Pressure, bar	155
Power Plant Thermal Efficiency	0.3311

Table 2. Reference TRU Isotopic Composition
(UO_2, 4.2 w/o U235, 50 MWd/kg Burnup, after 10 Years of Cooling)

Isotope	Weight %
U-234	0.0001
U-235	0.0023
U-236	0.0019
U-238	0.3247
Np-237	6.641
Pu-238	2.7490
Pu-239	48.6520
Pu-240	22.9800
Pu-241	6.9260
Pu-242	5.0330
Am-241	4.6540
Am-242m	0.0190
Am-243	1.4720
Cm-242	0.0000
Cm-243	0.0050
Cm-244	0.4960
Cm-245	0.0380
Cm-246	0.0060

The ORIGEN2 computer code [8] was used for the radioactive decay analysis of the fuel after the discharge from the reactor and for the evaluation of the spent fuel characteristics such as radiotoxicity, heat load and spontaneous fission neutron source.

The Doppler reactivity coefficient (DC), Moderator temperature coefficient (MTC), Void coefficient (VC) and Soluble Boron Worth (BW) were calculated for all considered fuel cycle options and assembly configurations at 3 time points: beginning (BOL), middle (MOL) and end (EOL) of fuel irradiation. In order to simulate close to realistic operating reactor conditions, all reactivity coefficients were calculated assuming that the soluble boron concentrations are 1000 ppm, 500 ppm, and 0 ppm at BOL, MOL, and EOL respectively.

The reactivity coefficients were calculated as follows.

$$MTC = \frac{\Delta K}{K_1 \times K_2 \times \Delta T_m} \tag{1}$$

where ΔT_m is the moderator temperature difference between two moderator temperatures T_1 and T_2 and K_1 and K_2 are infinite medium neutron multiplication factors corresponding to temperatures T_1 and T_2, respectively.

$$DC = \frac{\Delta K}{K_1 \times K_2 \times \Delta T_f} \tag{2}$$

where ΔT_f is fuel temperature difference between two fuel temperatures T_1 and T_2.

$$VC = \frac{\Delta K}{K_1 \times K_2 \times \Delta V} \tag{3}$$

where ΔV is the difference between two coolant void fractions V_1 and V_2.

$$BW = \frac{\Delta K}{K_1 \times K_2 \times \Delta C} \tag{4}$$

where ΔC is boron concentration difference in ppm.

It was assumed that all MAs in the fuel have the chemical form of $(MA)O_2$ with densities equal to the theoretical density of PuO_2.

In the fuel cycle scenarios with multi recycling of TRU, the efficiency of uranium separation was assumed to be 99.995%.

1.2 Double-Heterogeneous Effect Evaluation and Codes Benchmarking

The primary computational tool in this study was the CASMO4 2D lattice transport code widely used in the nuclear industry for the fuel management and core analysis of Light Water Reactors (LWRs). It produces very accurate results for the typical LWR fuel compositions and geometries. This part of the study has addressed two primary concerns

regarding the capabilities of CASMO4 to predict with a reasonable accuracy the criticality and nuclides evolution with the burnup for the fuel micro particles dispersed in the inert matrix.
The CASMO4 utilizes 70 energy groups cross-sections library that were generated using the typical LWR energy spectrum. In the fuel with large loadings of Pu and MA, the neutron energy spectrum tends to be much harder than generally encountered in a conventional LWR. Therefore, the cross-section library with the larger number of energy groups might be needed to produce accurate results for TRU containing fuel designs.

In addition, CASMO4 cannot explicitly handle heterogeneous structure of the fuel pellet. Therefore, only homogeneously mixed fuel and matrix materials in a solid fuel pellet can be modeled. The additional level of heterogeneous structure in the dispersed particle fuel creates an additional resonance self-shielding effect, which would be completely neglected in the CASMO4 calculations. The magnitude of this effect depends on the particle's size, composition and relative number densities of the matrix and the fuel.

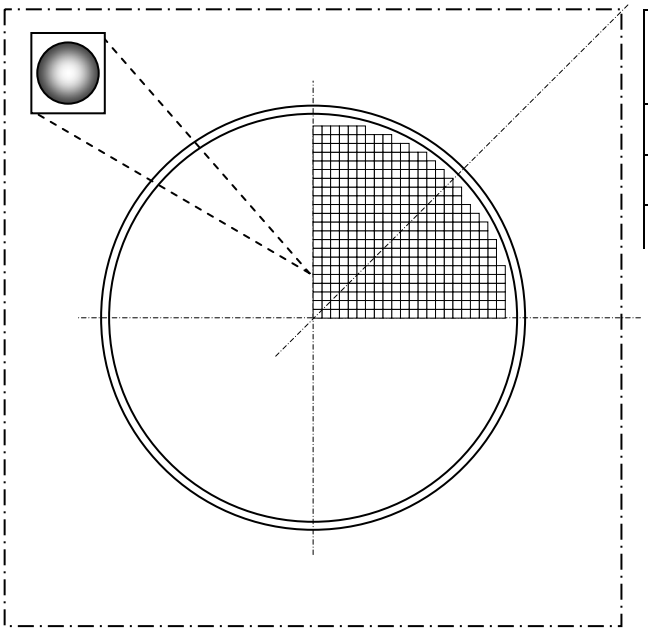
Three fuel pin cell burnup calculations were performed in order to assess the effect of using 70 group cross-section library and the effect of the double heterogeneous structure on criticality and the isotopes evolution predicted by the CASMO4 code.

In the first case, the fuel micro sphere geometry was explicitly modeled in MCNP4C [9]. The fuel pin cell geometry is schematically presented in Figure 1. The fuel micro particles were arranged in a simple cubic lattice with the total fraction of 30 v/o occupied by the fuel particles. All of the fuel particles had an identical diameter of about 150 μm. This particle size is close to the optimal one in terms of the mechanical and thermal properties of the fuel as well as its ability to sustain radiation damage [5],[10]. The burnup calculations were performed using the MCODE [11] – MCNP-ORIGEN linkage utility program. The ENDF-BVI based continuous energy cross-sections set was used for the MCNP calculations. The number of neutron histories in the Monte-Carlo simulation was chosen so that sufficient number of collision events had occurred in nearly every fuel

micro particle in order to ensure that the double-heterogeneous self-shielding effect is represented correctly.

In the second case, the MCODE calculation was repeated for the fuel pin cell of identical geometry and materials composition except for the fact that in this case the fuel particles (TRUs and YSZ) and the Spinel matrix were homogeneously smeared over the entire fuel pellet volume.

Finally, the homogeneously mixed fuel (TRU, YSZ and Spinel) case identical to the second one was calculated with CASMO4.



Fuel Particle: D = 150μ 50 v/o TRU O_2 50 v/o YSZ
Fuel Particles volume fraction in Spinel = 30 v/o
Cladding: Zr-4, 6.5 g/cm^3
Coolant: H_2O, 580K, 155 bar

Figure 1.Double-Heterogeneous Fuel Pin Cell Geometry

Figure 2 reports the results of the criticality prediction by MCODE and CASMO4 for the three calculated cases. The difference in k_∞ between the homogeneous and heterogeneous cases calculated with the MCODE is on the order of a fraction of a percent. At a number of data points the difference is larger than the statistical error. This suggests that the k_∞ value is somewhat higher in the heterogeneous case than is the homogeneous case although in general the overall double-heterogeneity effect is small. The small magnitude of this effect can be attributed to the small fuel particle size and relatively low concentrations of the fertile resonance nuclides in contrast to conventional UO_2 fuel with

the large concentration of U238. Another possible reason could be the fact that the total resonance absorption in fissile and fertile nuclides changes by a similar factor due to additional self-shielding introduced by the double-heterogeneous geometry so that the overall criticality of the system changes only marginally. The confirmation of these assumptions require additional investigation.

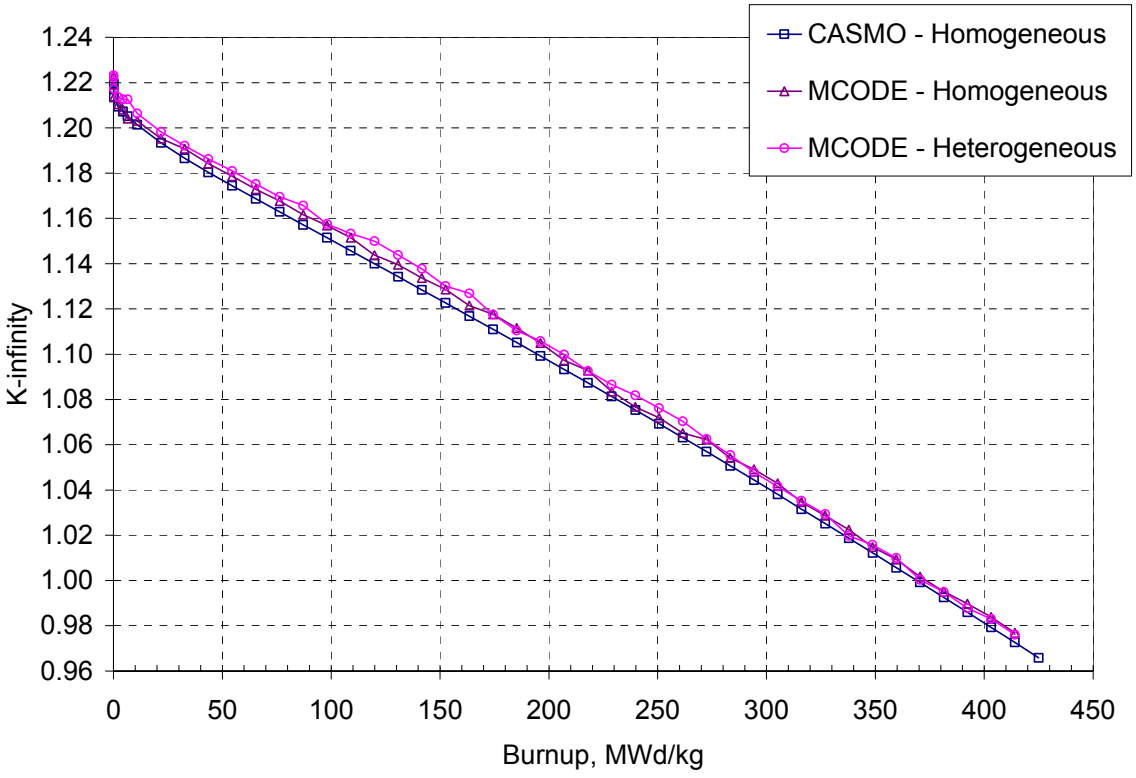


Figure 2. Criticality vs. Burnup for Homogeneous and Heterogeneous Geometries

In the homogeneous geometry, the difference between values predicted by CASMO4 and those by MCODE ranges from zero to about 0.5%. The beginning of irradiation k_∞ values agree very well but the difference increases with burnup up to about 100 MWd/kg then becoming rather constant up to the 400 MWd/kg. The larger k_∞ values predicted by MCODE can be mostly attributed to a discrepancy between the two codes in the prediction of Am242m evolution with burnup and due to a very large Am242m thermal fission cross-section (about 7000b). The Am242m number density as a function of burnup is shown in Figure 3. The Am242m number density calculated by CASMO4 is smaller than that calculated by the MCODE by the factor of about 1.7 at 150 MWd/kg. The Am242m builds up primarily from neutron captures in Am241. Since the Am241

number density changes with burnup are very similar for both codes (Figure 4), a possible reason for the discrepancy in Am242m buildup can be the incorrect treatment of the branching ratio between the metastable and ground state of Am242 in one of the codes or differences in cross-section libraries for Am242m.

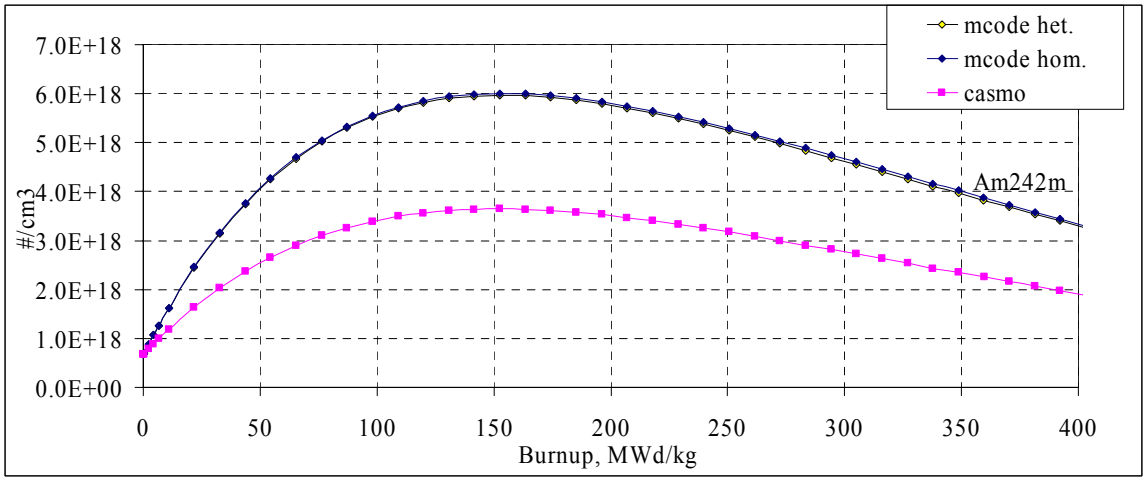


Figure 3. Am242m Number Density vs. Burnup

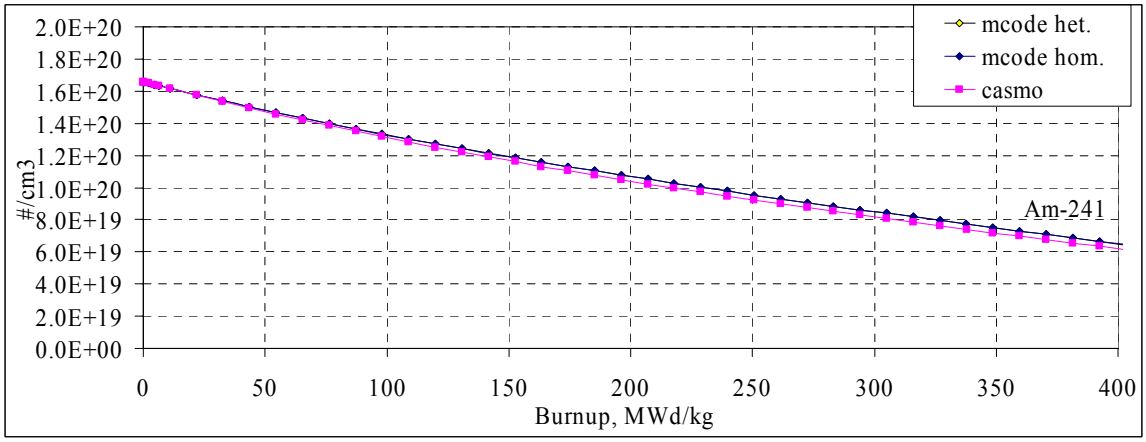


Figure 4. Am241 Number Density vs. Burnup

The predictions by the two codes of the number densities of all the Np, Pu and Cm isotopes are in reasonably good agreement. Minor discrepancies in the number densities prediction are most likely due to the differences in the cross-section data sets used. Selected results for the important actinides are summarized in an Appendix B.

In conclusion, the CASMO4 computer code can be used in scoping studies of the FFF designs with a reasonable degree of confidence. The treatment of Am242 branching ratio

in CASMO4 and cross-section library differences which probably lead to considerable discrepancies in predicted Am242m number densities require additional investigation. Otherwise, the predictions of criticality and TRU nuclides number densities by CASMO4 and MCODE are in a fairly good agreement.
No effect due to the limited cross-section library energy group structure was observed. The effect of the fuel micro particles homogenization can be considered as minor and is neglected for the purposes of this study.

1.3 Evaluation of Burn down Scenario

The main goal of the TRU "Burn down" fuel cycle option is to reduce the current stockpile of TRU from the existing LWRs in the most effective way. In order to achieve this goal, two primary objectives have to be attained; namely, the highest possible destruction rate and highest possible fractional burnup of TRU. In addition, substantial degradation of Pu vector to increase proliferation resistance of materials intended for final disposal in the repository is highly desirable.

In this part of the analysis, we compare two possible PWR core arrangements: homogeneous and heterogeneous. The homogeneous option refers to the reactor core with a single type of fuel assemblies. Each of the assemblies is composed of TRU fuel with the isotopic vector presented in Table 2 in combination with the fertile-free matrix. The heterogeneous reactor core includes two types of fuel assemblies. The fuel in the assemblies of the first type is composed of Pu only, while the fuel in the second type of assemblies is primarily composed of MA with some addition of Pu to sustain reasonable criticality constant of the assembly. This heterogeneous arrangement allows additional flexibility in optimization of TRU destruction efficiency. The Pu and Pu-MA containing assemblies can be optimized separately to burn efficiently the Pu and MA. In addition, the heterogeneous core allows separate fuel management schemes for the different types of assemblies. For example, if deeper MA burnup was found beneficial, the in-core residence time of the MA containing assemblies can be extended.

1.3.1 Homogeneous Option

The current PWR assembly geometry allows a certain degree of change for the optimization of the lattice moderator to fuel volume ratio (or Hydrogen to Heavy Metal ratio – H/HM). The H/HM ratio defines the fuel lattice energy spectrum and, therefore, affects the relative rates of destruction and generation of different TRU nuclides. A scoping study was performed to investigate the TRU destruction rate and fractional burnup sensitivity to the fuel lattice H/HM ratio and initial TRU loading.

The burnup calculations were performed in pin cell geometry. The H/HM ratio was varied in a wide range of values by changing the coolant density for the fixed pin cell geometry. As demonstrated in Reference [13], this approach of varying H/HM and other more realistic options, for example increasing the fuel pin cell pitch, can be considered as neutronically equivalent for the purposes of the current study.

The calculations were performed for 3 different initial TRU loadings of 10, 15 and 20 volume percent relative to the fuel pellet volume, which corresponds to 33.3, 50.0 and 66.7 v/o of TRU in the fuel micro particle volume respectively. The remaining volume fraction of the fuel micro particle was occupied by YSZ. The spinel matrix occupied a fixed (70 v/o) fraction of the fuel pellet volume. The choice of these three compositions was due to the following considerations. The 10 v/o of TRU composition represents a realistic reference case which results in approximately 18 months fuel cycle length in the reference PWR geometry. The 20 v/o of TRU is likely to be the limit of TRU loading from a materials behavior perspective as discussed in the Section II.

The results of the calculations are reported in Figures 5 through 8. The variation of H/HM ratio towards larger values results in a modest (up to 18%) increase in the reactivity limited burnup (Figure 5). The higher achievable burnup in turn leads to a more efficient destruction of TRU as demonstrated in Figure 6. Fuels with different TRU loadings have about the same optimal discharge burnup value for over-moderated lattices. As a result, the minimal residual fraction of TRU is approximately the same (~40%) for the fuels with different initial loadings. In the reference geometry and 10v/o TRU loading, which

provides discharge burnup corresponding to 18 month fuel cycle, about 53% of initial TRU can be destroyed per one fuel batch path through the reactor core.

The difference in the discharge burnup for different H/HM values has no effect on the destruction rate (Figure 7). For the FFF, the TRU destruction rate is determined solely by the core power. For a typical PWR core power density, the TRU destruction rate is about 1140 kg per GWe Year.

Although wetter than reference fuel lattices seem to be more attractive from a burnup viewpoint, for a fixed core volume and total power, an increase of H/HM ratio will result in a reduction of fuel volume and increase of power density in the fuel. Therefore, only moderate modifications in the fuel assembly geometry may be possible because of the thermal-hydraulics constraints. Alternatively, if the total core power is not fixed, satisfactory thermal-hydraulic design may be feasible by lowering the specific power in the fuel. However, this will result in considerable reduction in TRU destruction rate. Figure 8 illustrates that fact. The data shown in Figure 8 was obtained assuming that H/HM was changed through variation of the fuel pin cell pitch for the fuel rods of a fixed diameter.

The results of the calculations described above indicate that even in the reference PWR pin cell geometry the efficiency of TRU destruction is very close to the optimal one. This is a significant advantage because by using the reference assembly configuration, a near optimal burning efficiency can be achieved without impairing the destruction rate or changing the thermal-hydraulic design of the core.

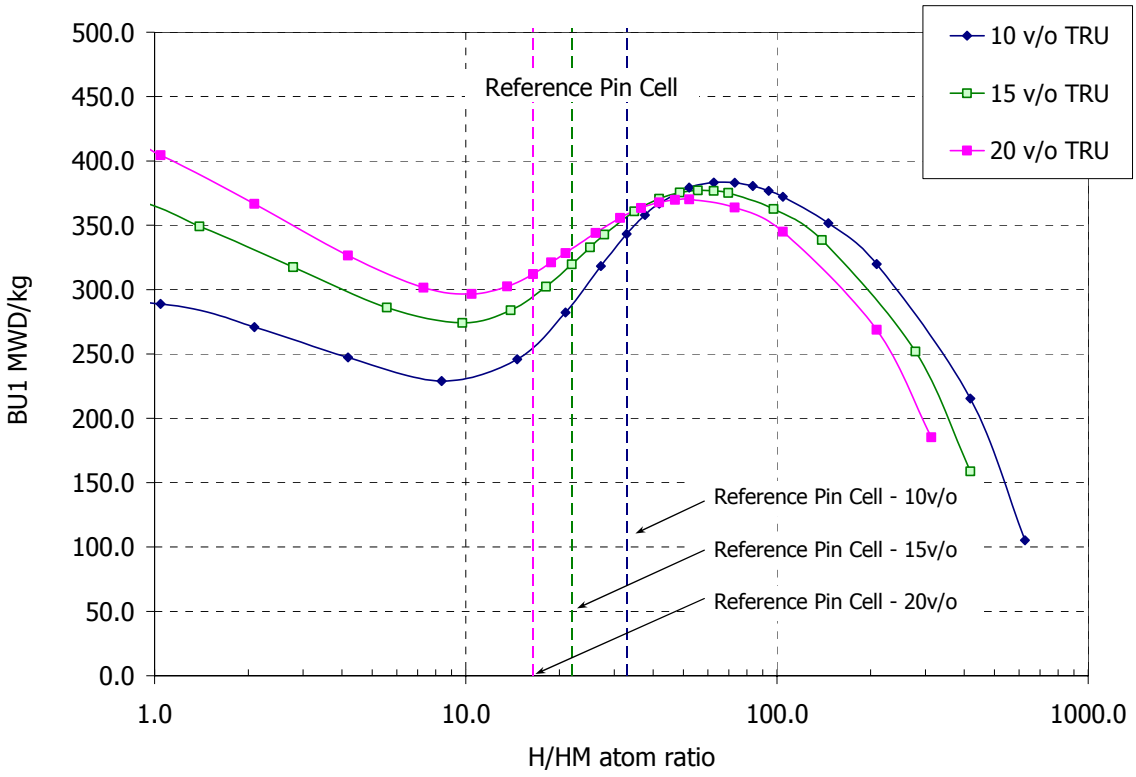


Figure 5. Single Batch Burnup vs. H/HM Ratio

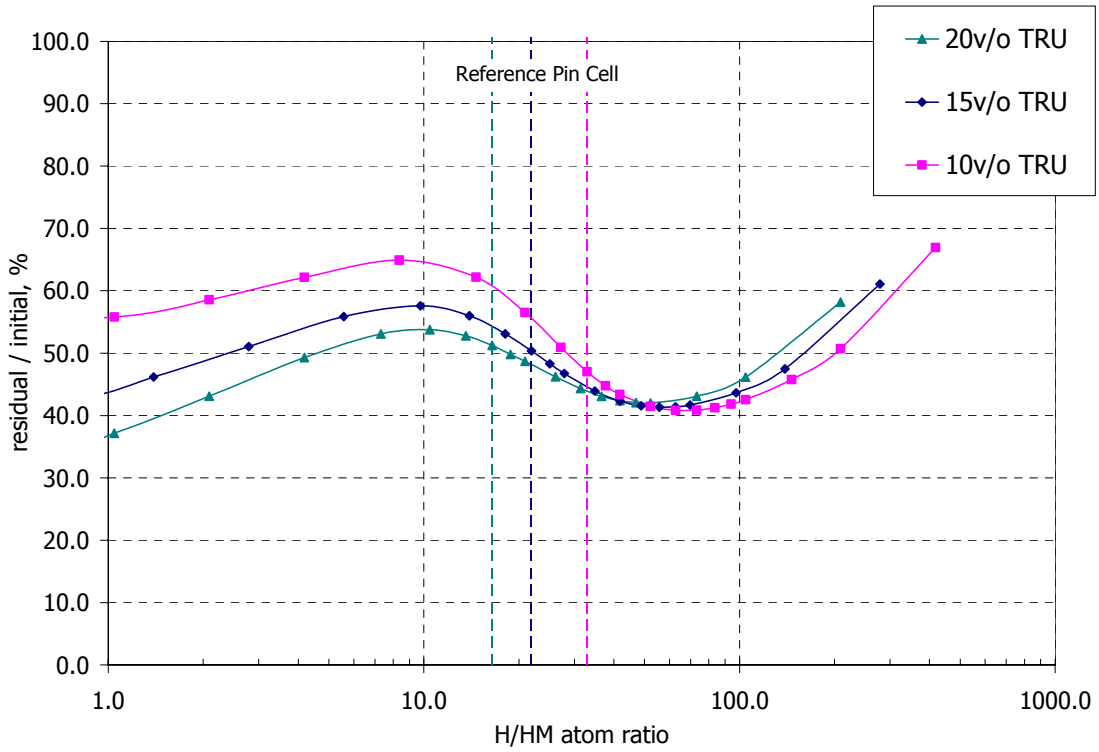


Figure 6. Residual Fractions of TRU in Discharged Fuel vs. H/HM Ratio

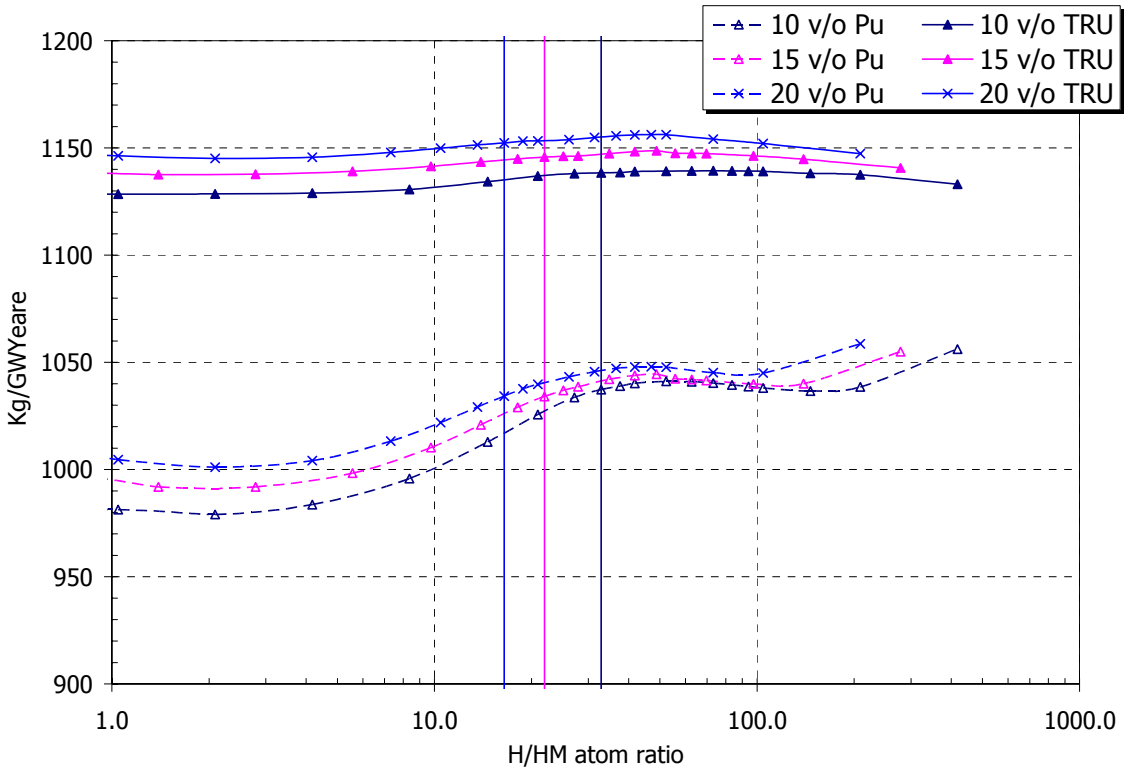


Figure 7. Energy Normalized Pu and TRU Destruction Rates vs. H/HM Ratio

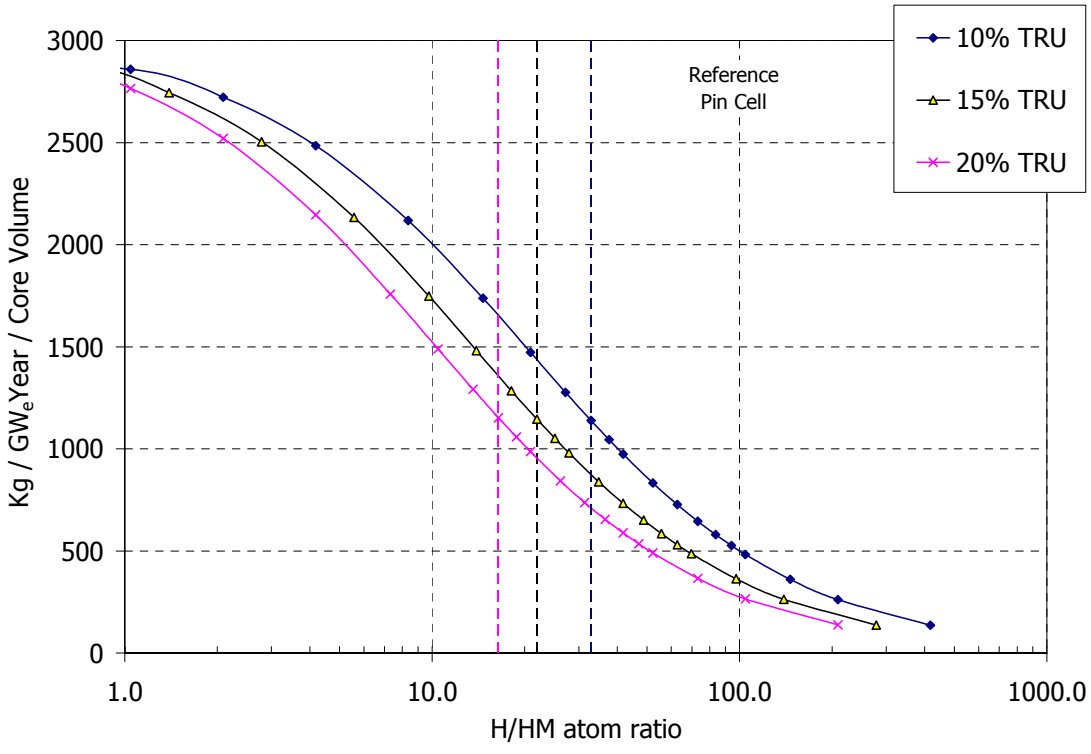


Figure 8. Pu and TRU Destruction Rates per Reference Core Volume

A reference PWR "assembly" burnup calculation with FFF containing 10 v/o of TRU was performed to assess the validity of the "pin cell" geometry calculations and to evaluate the possibility of an additional TRU recycling stage. In addition, the results of this calculation were used for evaluation and comparison of the spent fuel characteristics described in a later section. Finally, the reactivity feedback coefficients as well as the soluble boron worth for the reference fuel assembly geometry were also evaluated.

Burnable absorbers are likely to be used in a practical fuel design. Therefore, the burnable poison effect on the TRU destruction efficiency was also evaluated by calculation of a case with the addition of natural Er oxide to the fuel. The main advantage of Er as a burnable absorber is the presence of a large absorption resonance in Er167 that overlaps with the fission resonance of Pu239. As a result, Er can potentially improve the fuel temperature reactivity feedback (Doppler Coefficient). The main disadvantage of Er is a reactivity penalty at the fuel discharge point due to the absorption in some of the residual Er isotopes.

Figure 9 shows an example of criticality curves for the assembly calculations with and without burnable poison for the single once-through case and for the case with additional stage of TRU recycling. The composition of the once-recycled TRU fuel was chosen such that all the unburned TRUs from the first stage (after the fission products separation) are included and enough "fresh" TRU is added to be able to reach the first stage fuel cycle length.

Table 4 summarizes the fractional TRU burnup for the poisoned and un-poisoned cases, with and without TRU recycling. The effect of BP addition is important since BP considerably reduces TRU burning efficiency due to the residual reactivity penalty and loss of neutrons to BP that otherwise could be used for transmutation and fission of TRU.

Degradation of TRU isotopic vector after the 1st path makes TRU recycling unattractive because of considerably lower fractional burnup in the case where minimal reprocessing is the first priority. However, if multiple reprocessing is not of a great concern, an

equilibrium fuel composition can be achieved after several recycles with the fractional TRU burnup converging to values between 25% and 30%. In that case, the entire TRU stockpile can theoretically be destroyed.

Pu isotopic composition after once-through TRU burning is also degraded to a considerable extent which substantially enhances proliferation resistance of the spent fuel. The final Pu isotopic composition in the discharged FFF is shown in Table 4.

Table 3. TRU Destruction in Homogeneous FFF Core:

Normalized Material Flow Summary

Case	1st path		2nd path	
	No BP	With BP	No BP	With BP
Discharge Burnup, MWD/kg	541	486	334	292
TRU Loaded, kg / GWeY	2037	2264	3295	3774
TRU Discharged, kg / GWeY	887	1112	2122	2594
Pu Loaded, kg / GWeY	1759	1954	2744	3137
Pu Discharged, kg / GWeY	671	850	1623	1996
% TRU Burned / path	56.5	50.9	35.6	31.3
% Pu Burned / path	61.8	56.5	40.8	36.4

Table 4. Initial and Discharge Pu Isotopic Composition.

Pu Isotope	w/o	
	Initial	At discharge
238	3.18	17.23
239	56.35	9.77
240	26.61	31.36
241	8.02	19.49
242	5.83	22.15

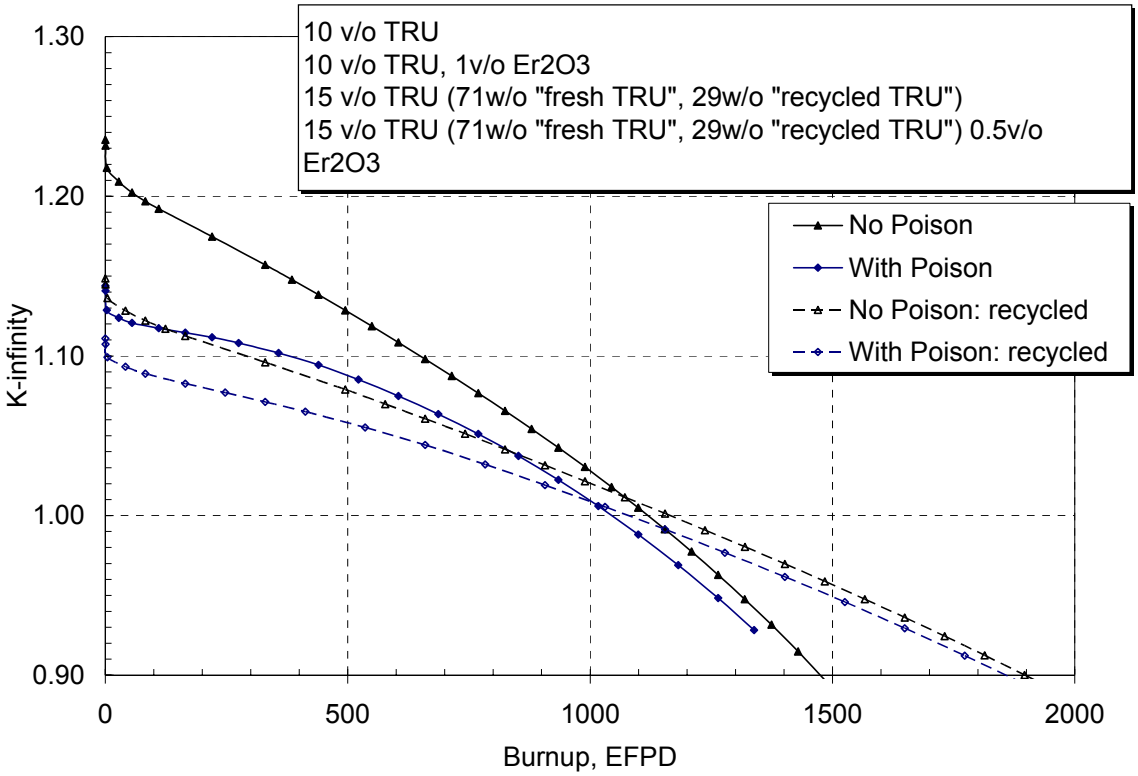


Figure 9. Criticality Curves for the FFF Assembly Cases

1.3.2 Macro-Heterogeneous Option

Heterogeneous core configurations allow additional flexibility in the optimization of TRU destruction efficiency.

In this part of the work, we examine the effect of variation of the relative amounts of Pu and MA in fuel assemblies as well as of the assembly lattice H/HM ratio on burning efficiency. Assuming that Pu and MA come in different streams from the chemical separation process, the relative amounts of Pu and MA can be changed so that most of the Pu is concentrated in one type of fuel assemblies, while all of the MA are concentrated in the second type of the assemblies.

The destruction rate of TRU in FFF is determined only by the specific power in the core. Therefore, the main objective was an increase in reactivity limited burnup, which leads to a higher degree of TRU destruction. An additional goal was the investigation of the possibility to accelerate MA destruction rate at the expense of Pu destruction rate.

The heterogeneous equilibrium core performance was modeled using the CASMO4 "colorset" option. This option allows 2D transport calculations with burnup for a 2x2 segment of different full size fuel assemblies (Figure 10).

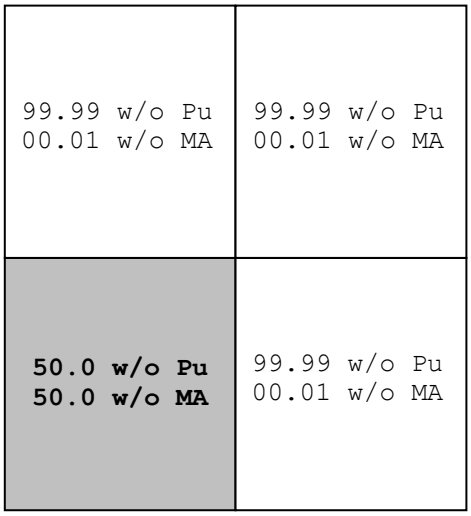


Figure 10. Example of CASMO Colorset Layout

A systematic analysis of different heterogeneous assembly configurations was performed by varying separately the composition and H/HM of the two assembly types. Several constraints were imposed on the optimization to ensure the feasibility of a realistic design:

- Colorset pin power peak ratio < 1.2.
- TRU volume fraction in the fuel particles < 70 v/o (< 20 v/o of the pellet volume).
- Variation in the fuel pin diameter < ≤ 20%.
- Colorset average Pu to MA ratio is conserved and equal to the one used in the homogeneous assembly evaluation (Table 2).

The results of this study show that no significant improvement in heterogeneous colorset burnup can be achieved in comparison with the homogeneously mixed Pu and MA

assembly. Moreover, the rate of MA destruction in the heterogeneous configurations is always lower than in the homogeneous assembly under the imposed constraints.

Selected results comparing the homogeneous and heterogeneous options are reported in Table 5. In Table 5, the heterogeneous case 2 denotes a colorset with the lattice geometry identical to the homogeneous assembly case. The Pu and MA containing assembly lattice H/HM ratio was varied in a range of ≤20 % of the reference one. The heterogeneous cases 1 and 3 (Table 5) are the best performing cases in terms of reactivity limited burnup and MA destruction rate respectively. In the case 1, the Pu containing assembly has higher than reference H/HM by 20% and the Pu-MA assembly has lower than reference H/HM by 20%, while in the case 3, the Pu containing assembly has lower than reference H/HM by 20% and the Pu-MA assembly has higher than reference H/HM by 20%.

Table 5. Efficiency of TRU Destruction: Homogeneous vs. Heterogeneous Option

Case	Burned as % of Initial			Kg Burnt / GWeY		
	TRU	Pu	MA	TRU	Pu	MA
Homogeneous	56.3	61.6	23.0	1135	1072	63
Heterogeneous, (case1)	56.4	62.5	16.1	1134	1090	44
Heterogeneous, (case2)	55.4	61.5	16.4	1135	1089	46
Heterogeneous, (case3)	54.3	60.2	16.6	1135	1088	48

Although the heterogeneous option is not advantageous in terms of the TRU destruction efficiency in comparison with the homogeneous option, the heterogeneous core configuration allows different fuel management schemes for different assembly types. As a result, degree of MA burnup can be improved by extending the MA assemblies residence time. The MA burning assembly can be designed to have a very flat reactivity over the entire irradiation time. Figure 11 shows an example of criticality versus burnup curves for a number of possible compositions of the MA burning assembly. The flat

reactivity ensures limited impact on the power peaking in any heterogeneous core. However, the homogeneous option may be preferable because better transmutation efficiency can be achieved and because Pu and MA do not require separation during reprocessing.

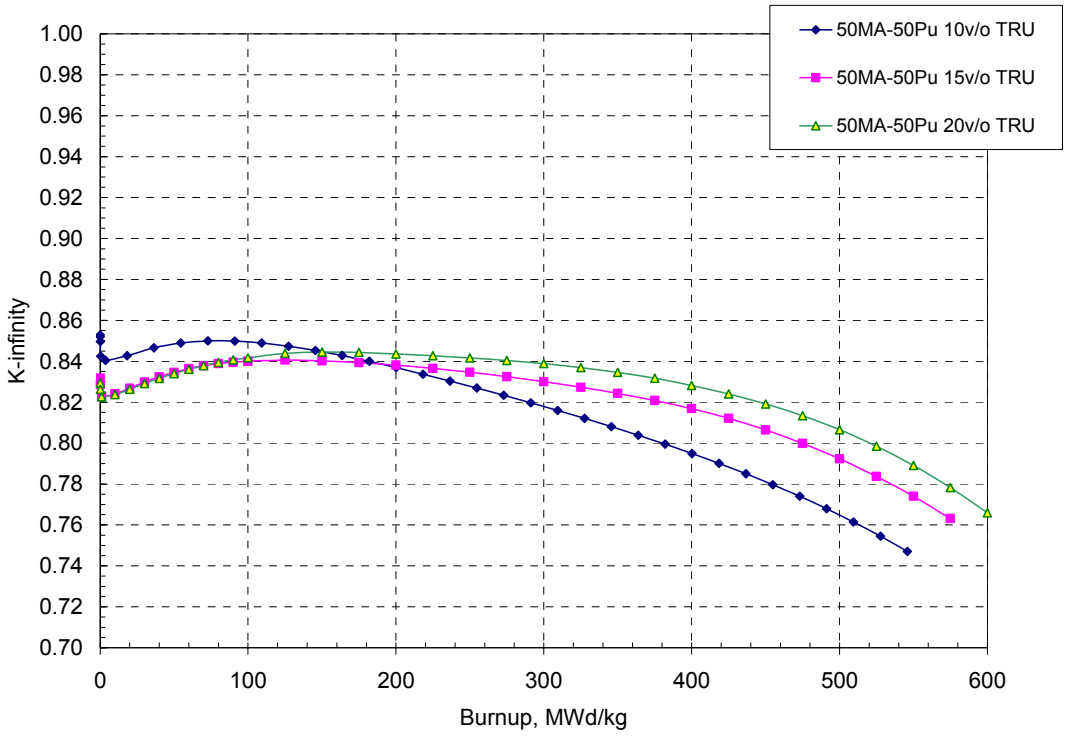


Figure 11. Criticality of MA Burning Assembly

1.3.3 Spent fuel Characteristics

One of the major objectives for the development of any TRU transmutation concept is the reduction of potential spent nuclear fuel impact on the environment. This section will compare the different options described earlier in terms of the following spent fuel characteristics.

- Radioactivity.
- Decay heat thermal power.
- Ingestion radiotoxicity.

- Spontaneous fission and (α,n) reaction neutron source.

The calculations were performed using the ORIGEN2.2[8] code with the library of radiotoxicity coefficients in units of Sv/Bq from ICRP Publication 68 [14].
Figures 12 through 15 compare the spent fuel characteristics normalized per 1 GWeYear of energy produced by the fuel for the homogeneous and heterogeneous FFF assemblies, for the typical MOX assembly (7 w/o of reactor grade Pu, 93 w/o of natural uranium) and for the reference all-uranium fuel. Solid lines in Figures 12-15 indicate actinides contribution only, while dotted lines designate the total spent fuel characteristic quantity from actinides and fission products. About 200 fission product isotopes provided in the CASMO4 libraries were included in the analysis.

The short term radioactivity and heat load are primarily determined by the fission products and are comparable for MOX fuel and FFF. The spontaneous fission neutron source (SFS) is considerably higher for FFF than for MOX up to 1 million years timeframe due to buildup of the higher MA and especially Cm isotopes. The SFS is an important characteristic because it can complicate the TRU separation and fabrication process in case TRU recycling is considered.

The activity, heat generation and radiotoxicity are higher for the FFF than for MOX fuel in the short term but become slightly more favorable in the time interval between 10^3 and 10^4 years. In the interval between 10^4 and 10^6 years, FFF and MOX fuel have comparable characteristics.

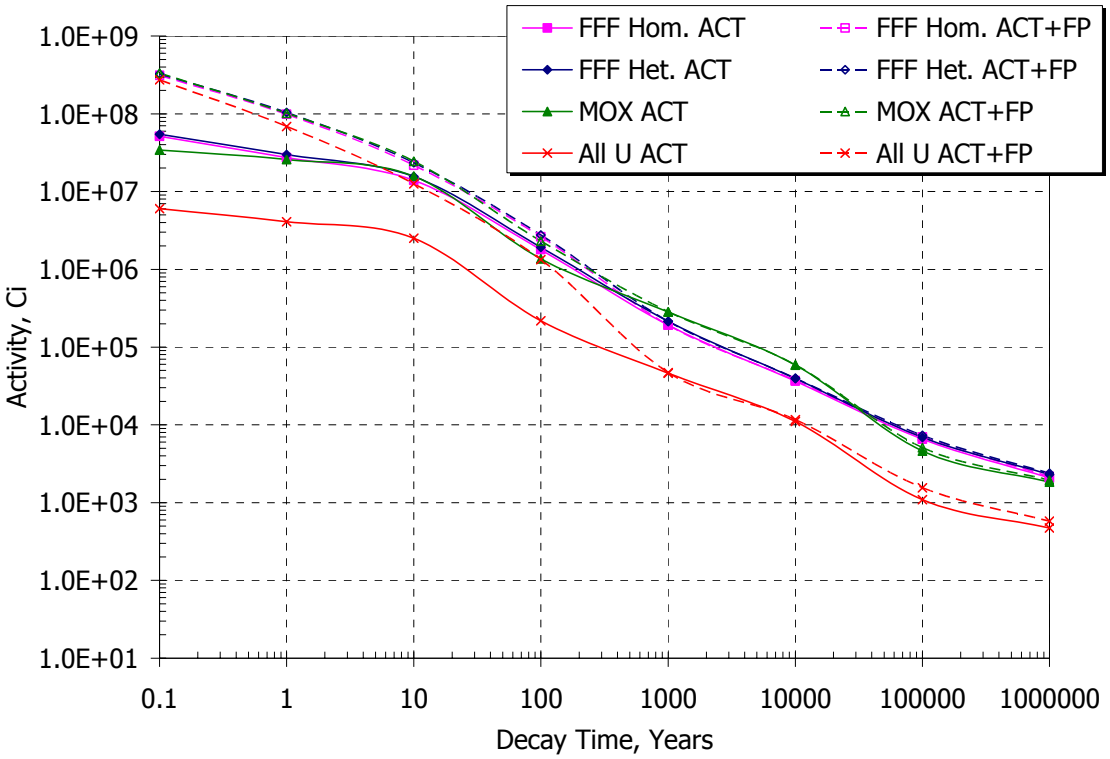


Figure 12. Radioactivity Normalized per 1 GWe Year of Last Core

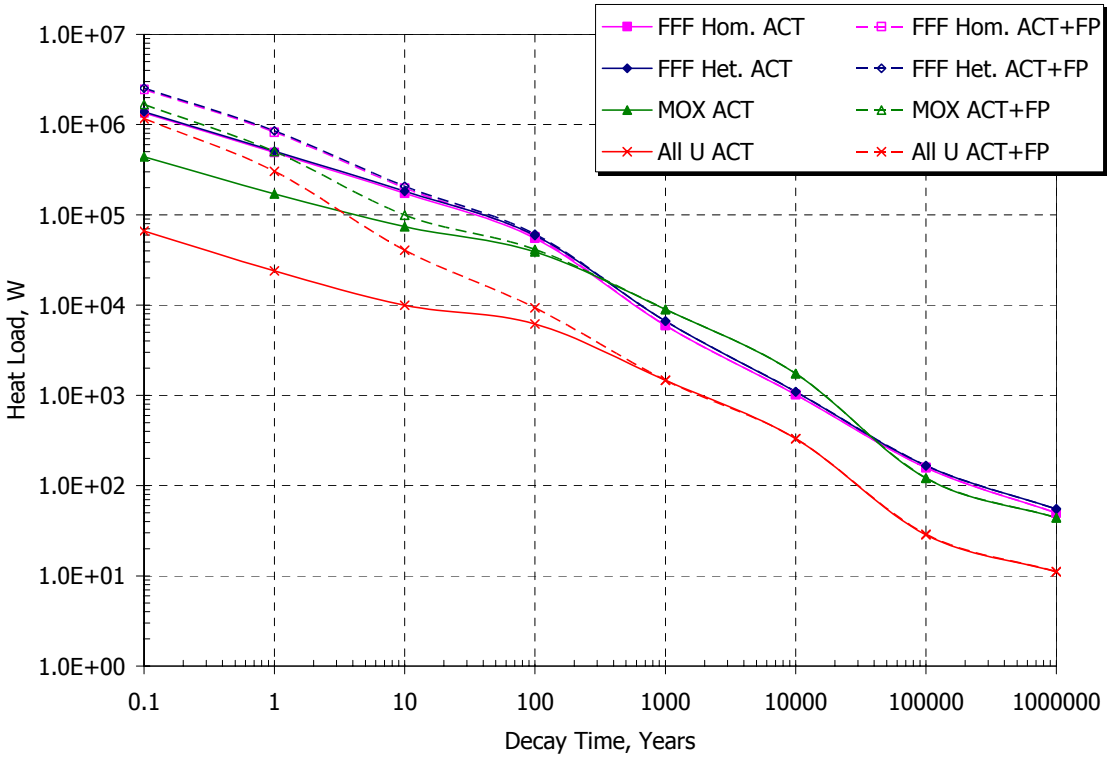


Figure 13. Decay Thermal Power Normalized per 1 GWe Year of Last Core

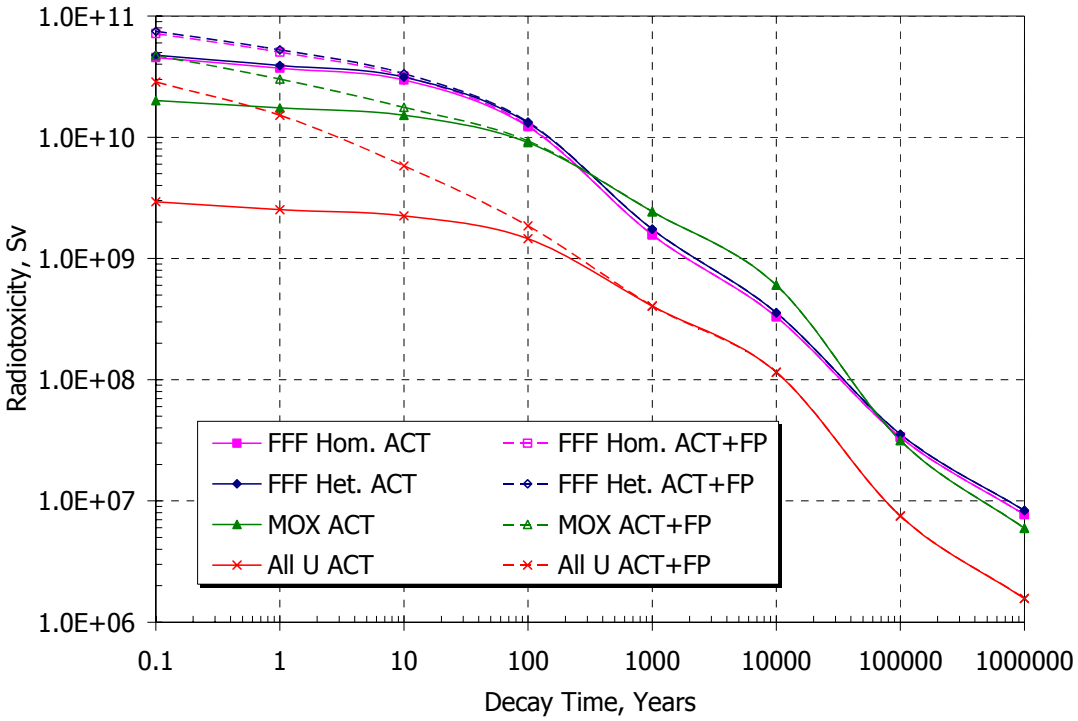


Figure 14. Ingestion Radiotoxicity Normalized per 1 GWe Year of Last Core

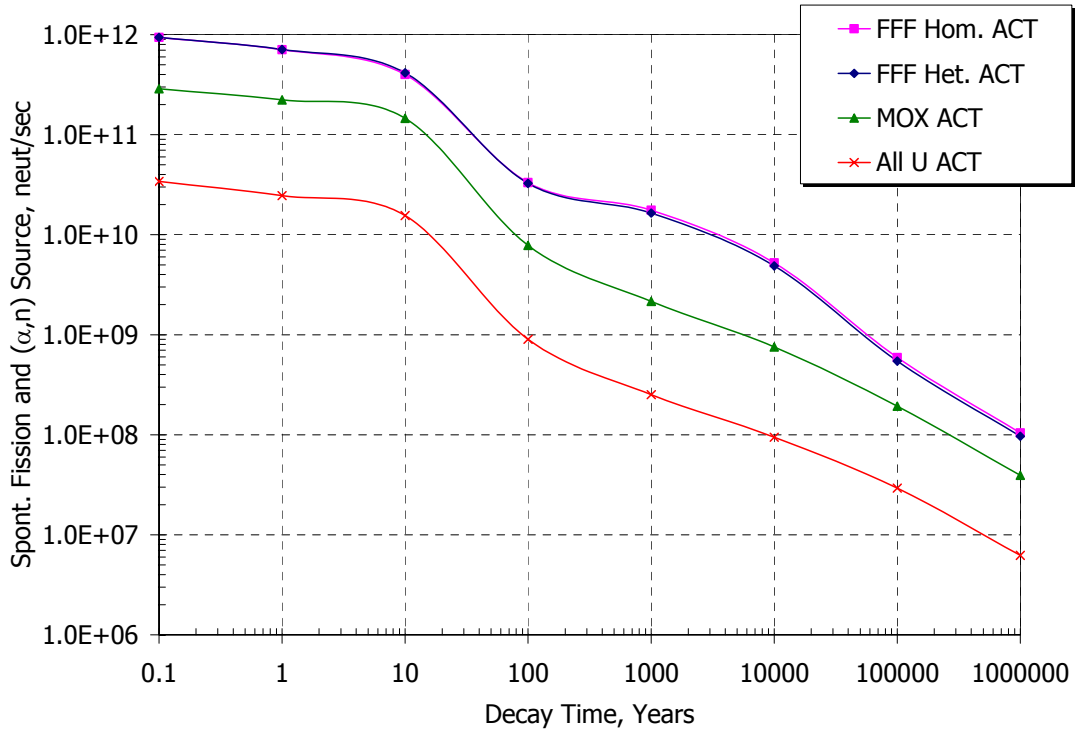


Figure 15. Total SF and (α,n) Neutron Source Normalized per 1 GWe Year of Last Core

The all-U fuel has the most favorable spent fuel characteristics among the all analyzed options and over the entire decay time interval considered. However, it is important to mention that for the data presented in Figures 12-15, only the energy produced by the fuel during its irradiation was accounted for. This fact results in somewhat inconsistent comparison of all-U spent fuel and the different options for its recycling. For example, the initial loading of one FFF assembly contains recycled TRU from more than 8 spent all-U fuel assemblies. Furthermore, this FFF assembly destroys about one half of loaded TRU while also producing energy. Therefore, in order to make a consistent comparison, the spent FFF assembly characteristics have to be re-normalized per total energy produced by the FFF assembly and recycled all-U assemblies that provided TRU for this FFF assembly.

Figures 16 through 19 present a comparison between the all-U fuel disposed directly and the all-U fuel that was recycled and burned in FFF assembly normalized per total energy produced by the fuel in all-U and FFF assemblies. Figures 16-19 show only the actinides contribution to the spent fuel characteristic quantities.

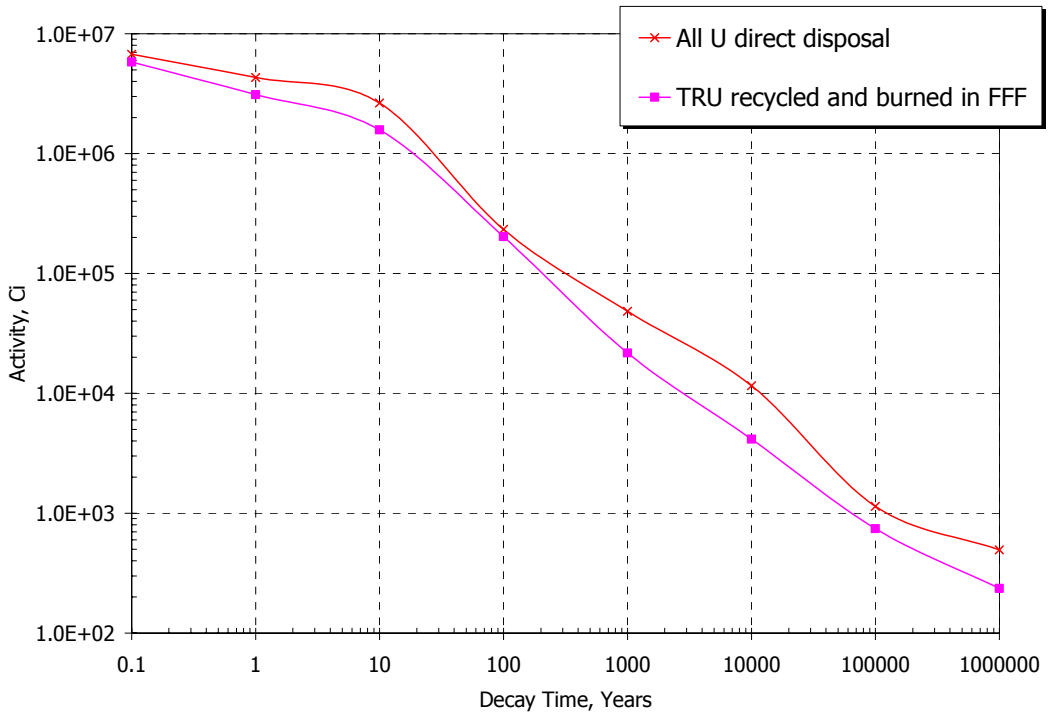


Figure 16. Actinide Radioactivity Normalized per 1 GWe Year of Total System Energy

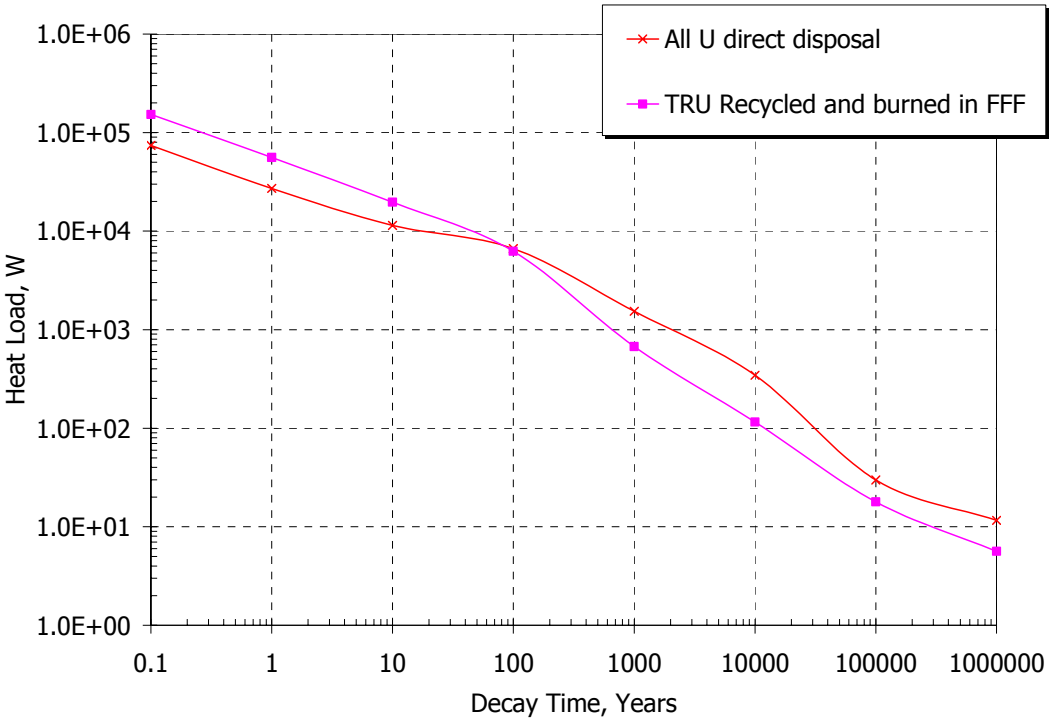


Figure 17. Actinide Decay Thermal Power Normalized per 1 GWe Year of Total System Energy

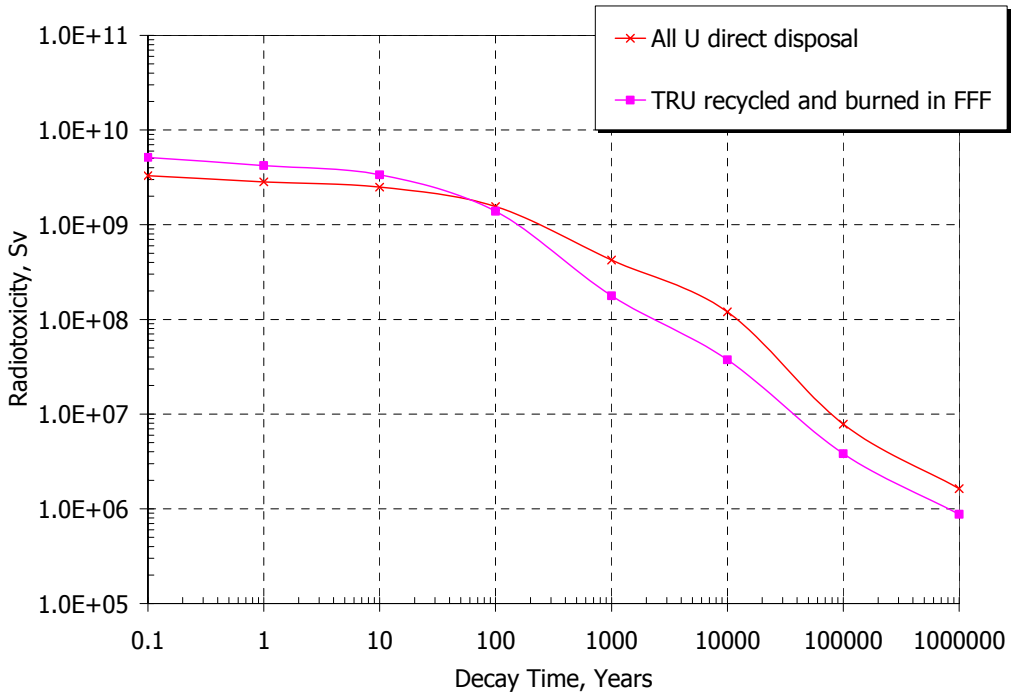


Figure 18. Actinide Ingestion Radiotoxicity Normalized per 1 GWe Year of Total System Energy

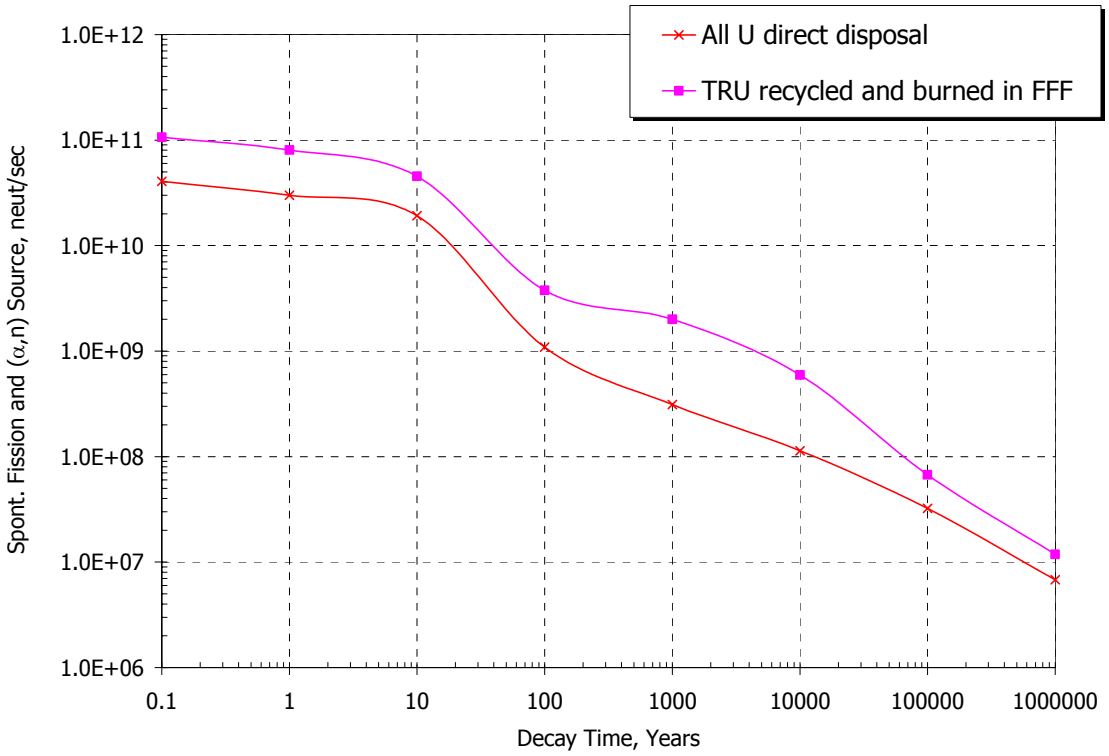


Figure 19. Actinide Total SF and (α,n) Neutron Source Normalized per 1 GWe Year of Total System Energy

The consistent comparison of FFF and all-U fuel indicate that the TRU recycling can moderately reduce the mid and long term (from 100 to 10^6 years) radioactivity, heat generation and radiotoxicity. The short term characteristics are comparable for the recycled and directly disposed TRU.

In conclusion, the analysis of the spent fuel characteristics has shown that the one stage recycling of TRU from a conventional LWR spent fuel and subsequent burning in FFF can reduce the amount of TRU by the factor of two. However, the impact of the one stage TRU recycling on the spent fuel characteristics is limited. These are preliminary conclusions which will be further examined in future studies to reflect the effect of the constraints assumed here.

1.3.4 Reactivity and Control Characteristics

All reactivity coefficients were calculated on a fuel assembly basis. Core average reactivity coefficients would be somewhat different as a core is composed of fuel assemblies with different accumulated burnup. However, the assembly based calculations can be used for comparison of FFF against reference UO_2 and typical MOX fuel evaluated on the same basis.

The results of reactivity coefficient as well as soluble boron worth calculations are summarized in Table 6. Only the homogeneous FFF option was evaluated since the heterogeneous core arrangement does not offer any significant advantages from the TRU destruction efficiency perspective.

The FFF assembly has a much lower value of DC although its value is still negative. The presence of Er burnable poison does not improve DC significantly.

The soluble boron worth (BW) is considerably lower for FFF in comparison with All-U fuel, however, it is comparable with MOX fuel. The lower BW for the FFF is due to the much harder (than in PWR) neutron spectrum as consequence of a strong thermal neutrons absorption in Pu and MA isotopes.

The MTC and VC are also negative at BOL with the soluble boron concentration of 1000ppm. Realistic values for MTC and VC can be obtained only with a whole core simulation with reasonable burnable poison design. In that case, the amount of excess reactivity to control and soluble boron worth will determine the realistic boron concentration at which then MTC and VC can be evaluated.

In summary, the results obtained do not indicate any significant FFF implementation problem related to reactivity feedback coefficients. Compared to a reference PWR, the much smaller soluble boron worth, which is common for Pu and MA containing fuels, is likely to impose additional requirements on the reactor reactivity control design features.

Table 6. Reactivity Coefficients and Soluble Boron Worth

		FFF 1st path		MOX	All-U
		No Poison	with 2%Er		
DC	BOL	-0.63	-0.75	-2.73	-2.03
	MOL	-0.77	-0.77	-2.87	-2.65
	EOL	-1.04	-1.08	-3.03	-3.08
MTC	BOL	-21.88	-32.89	-42.48	-11.26
	MOL	-32.39	-41.27	-54.36	-39.54
	EOL	-51.75	-61.63	-73.98	-66.29
VC	BOL	-54.07	-80.94	-112.48	-34.76
	MOL	-92.02	-114.92	-148.78	-119.03
	EOL	-172.04	-197.64	-204.00	-205.98
BW	BOL	-2.34	-2.51	-2.70	-6.11
	MOL	-3.42	-3.46	-3.17	-7.09
	EOL	-8.02	-8.10	-3.91	-9.51

Another important fuel characteristic which directly affects the feasibility of reactor control is the effective delayed neutrons fraction (β_{eff}).

Figure 20 reports the effective delayed neutron fraction for FFF assembly as a function of burnup for the 1st and 2nd TRU burn down path as well as for the reference PWR and typical MOX fuel. The β_{eff} values for FFF at the beginning of irradiation are lower than 0.003. Although β_{eff} increases monotonically with burnup due to increasing contribution to the total power from Pu241 fissions and decreasing contribution of Pu239 fissions, relatively low initial value is likely to impose a major limitation on the feasibility of PWR core with 100% loading of TRU in FFF assemblies.

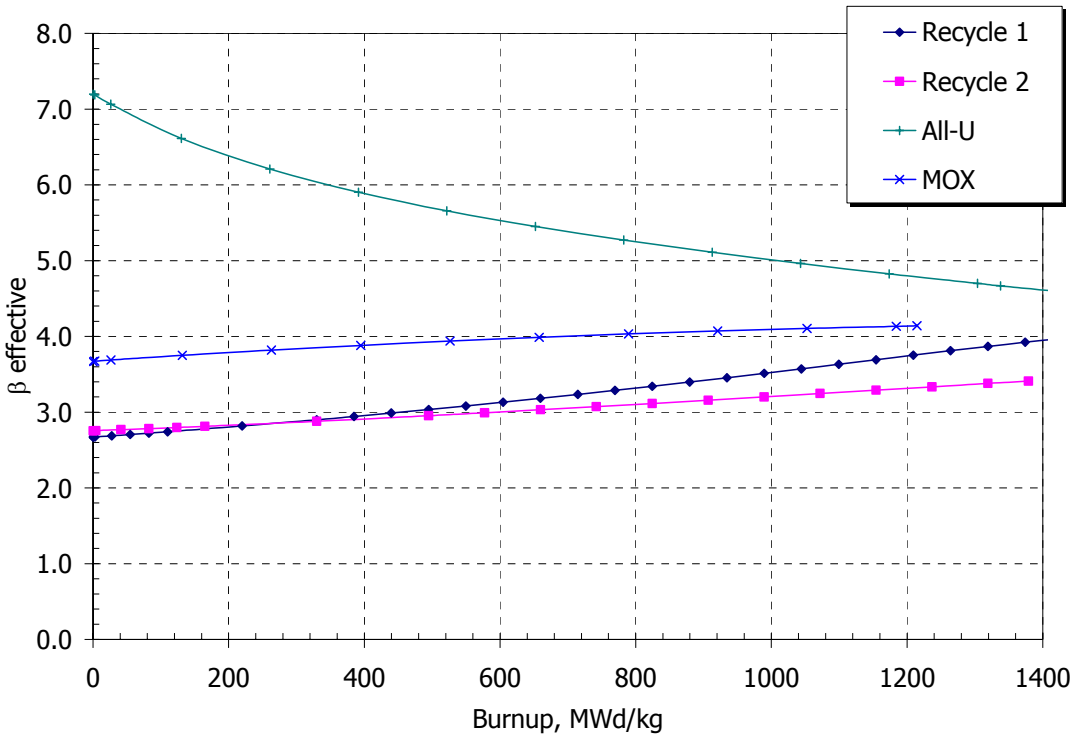


Figure 20. Effective Delayed Neutron Fraction × 10^3 vs. Burnup

1.4 Evaluation of Sustainable Fuel Cycle Scenario

This part of the study is aiming at the development and assessment of a fuel concept that will allow a complete recycling of TRU in conventional PWR fuel cycle. The fuel assembly design evaluated in this study suggests displacing some of the UO_2 fuel pins in conventional PWR assembly with fertile free fuel pins (Figure 21). Further in the text, this concept will be denoted as Combined Non-fertile and Uranium (CONFU) assembly. Each time such a CONFU assembly is discharged from the core, the residual TRU from FFF pins and the TRU that were generated in UO_2 pins are separated and prefabricated into a new CONFU assembly with "fresh" 4.2% enriched uranium pins and FFF pins that include all the TRU from the previous cycle.

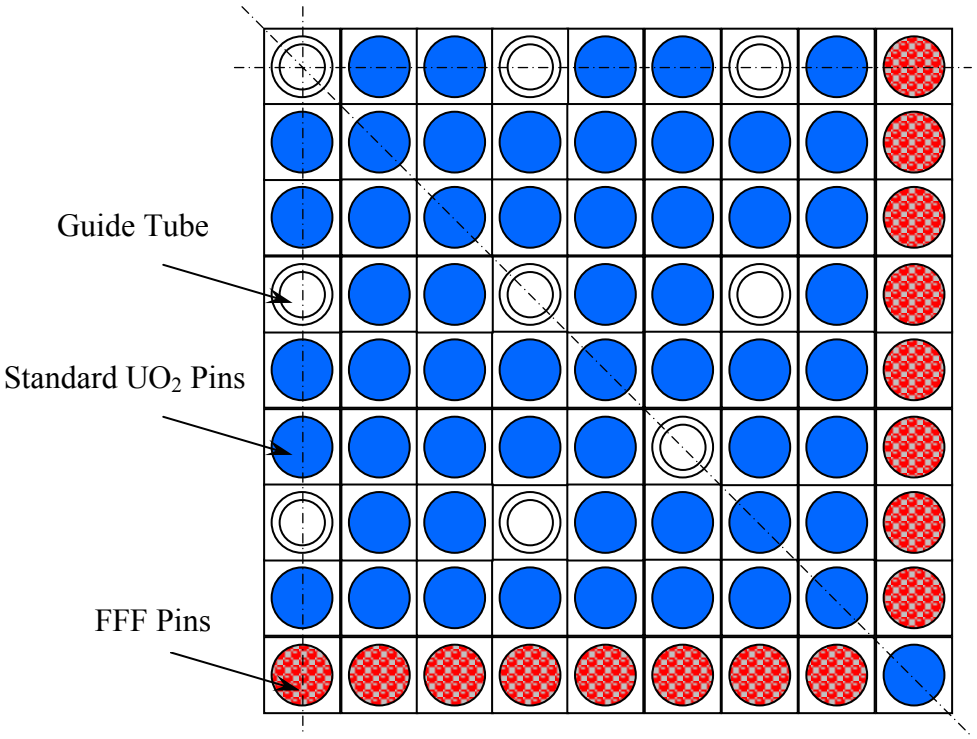


Figure 21. CONFU Assembly Configuration

The main objective of this part of the study was to evaluate the CONFU assembly concept with respect to its ability to achieve an equilibrium state with zero net generation of TRU and a constant fuel cycle length. In addition, similar to the evaluation of the TRU Burn down scenario, the reactivity feedback coefficients as well as selected waste stream characteristics were evaluated.

The number of FFF pins, initial loading of TRU and locations of FFF pins in the assembly were adjusted in order to achieve an equilibrium state with zero net balance of TRU generation and destruction.

The results of the preliminary evaluation show that between 52 to 60 FFF pins per reference 17×17 PWR fuel assembly is sufficient to attain an equilibrium TRU balance keeping TRU volume fraction in the fuel micro particles between 40 and 50 v/o (or between 12 and 15 v/o of the total fuel pellet volume)

The location of the FFF pins has a significant effect on power in the FFF pins and, therefore, affects TRU destruction efficiency. Figure 21 shows several examples of the BOL pin-power maps for candidate CONFU assembly configurations and illustrates the effect of FFF pins location on their power. The FFF pins generally have higher power when surrounded by UO_2 pins or have extra coolant (guide tube) in their vicinity. For such configurations, the power in the FFF pins is too high to satisfy thermal-hydraulic limits. Grouping the FFF pins together and addition of burnable poison (natural Er_2O_3) were explored as possible strategies to reduce pin peak power to acceptable values. The power map shown in Figure 22 at the right bottom corner (Case4) represents possible CONFU assembly configuration with reasonable power peaking factor of 1.25. In this particular case, 2 v/o of Er_2O_3 was added to FFF pins composition. The power map at the left bottom corner in Figure 22 (Case 3) represents another possible CONFU assembly configuration without employing any burnable poison.

The minimum DNBR calculations were performed to ensure the feasibility of CONFU assembly thermal-hydraulic design. Detailed single assembly modeling was done using the VIPRE-01 [15] sub-channel analysis code which is widely used for the evaluation of PWR thermal-hydraulics performance. The following assumptions were made to ensure conservative results: 18% core overpower, 294.7 ºC inlet coolant temperature, 1.56 assembly to core average power peak, “chopped” cosine axial power profile with peak to average ratio of 1.4.

The MDNBR value obtained for the un-poisoned CONFU assembly configuration (Case 3 in Figure 22) using W-3L correlation with l-grids is 1.721 which indicates that the concept can potentially have a sufficient thermal margin.

Consequently, case 3 and case 4 shown in Figure 22 were chosen for further analysis as representative candidate configurations of un-poisoned and poisoned CONFU assemblies with acceptable thermal-hydraulic performance.

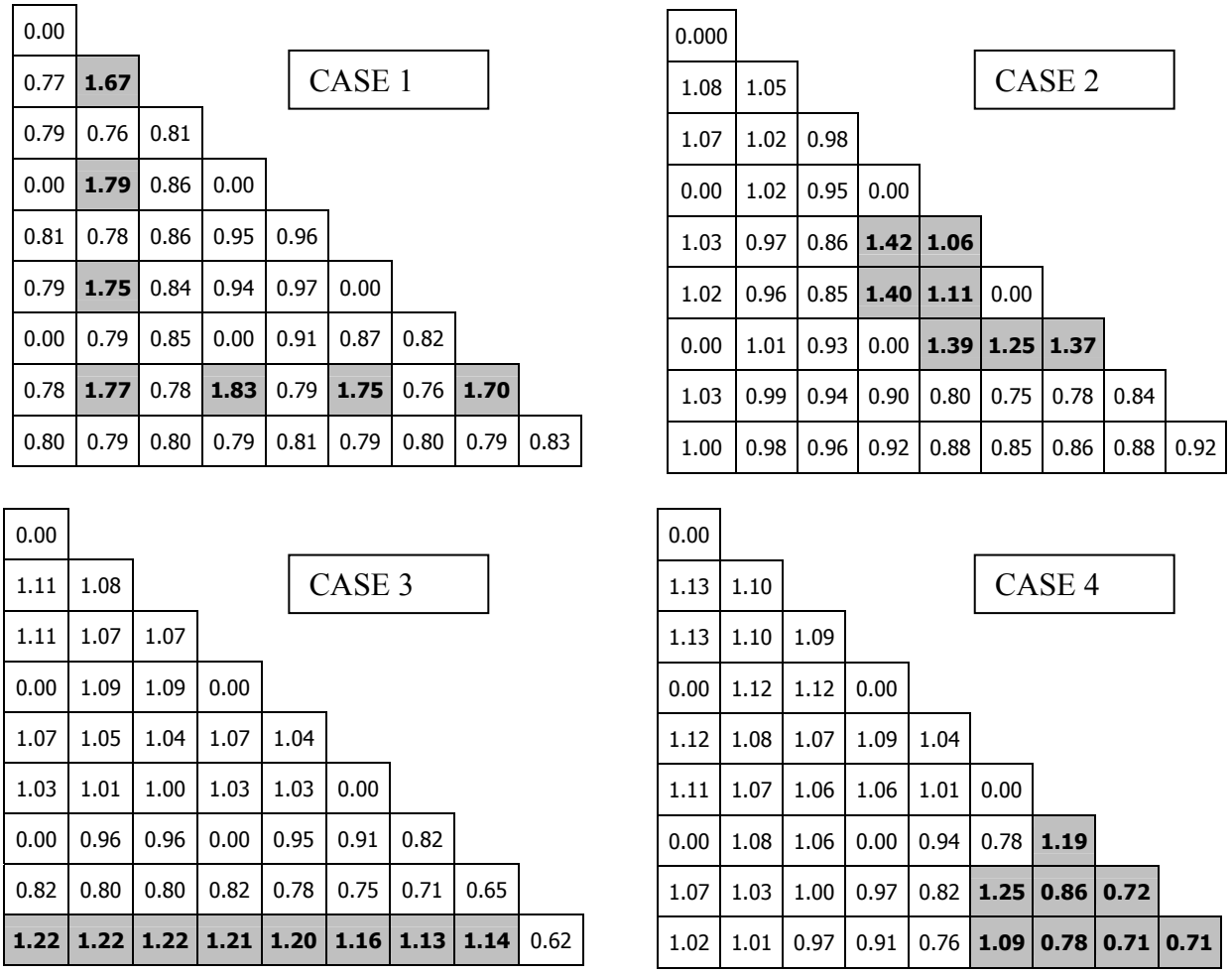


Figure 22. CONFU Assembly BOL Pin Power Distribution: Selected Design Options and Results

Five subsequent poisoned and un-poisoned CONFU assembly (cases 3 and 4 respectively) cycles were modeled to assess the possibility of a sustainable scenario from the net TRU balance viewpoint. Five years of cooling time was assumed between the fuel discharge and its reprocessing. Additional 2 years were allowed for the fuel handling and fabrication. The number of FFF pins per assembly and the amount of TRU loaded each cycle was conserved. In this case, the convergence of fuel cycle length to some constant value and stabilization of TRU composition would be the primary indicators of an approach to the equilibrium state.

The presence of burnable poison in the FFF pins significantly impairs the efficiency of TRU destruction (Table 7) due to the lower power in FFF pins and competition for neutron absorption between TRU nuclides and the burnable poison. Therefore, higher initial TRU loading is required for the CONFU assembly with burnable poison to achieve the equilibrium. Table 8 and Table 9 summarize the cycle-by-cycle fuel materials flows for the un-poisoned and poisoned CONFU assemblies respectively.

Table 7. TRU Destruction Efficiency for FFF Pins in CONFU Assembly

TRU Burnup, %	Recycle Stage				
	1	2	3	4	5
Without BP	50.78	40.13	37.32	36.21	34.63
With BP	32.62	30.60	27.94	26.96	26.60

Table 8. Un-Poisoned CONFU Assembly: Materials Flow Summary (per 1 GWeY)

	Recycle Stage					
	1	2	3	4	5	6
Total HM Loaded, kg	16,149	18,698	19,330	19,470	19,538	19,582
Uranium Loaded, kg	15,578	18,037	18,647	18,781	18,847	18,889
TRU Loaded to FFF pins, kg	580	671	694	699	701	703
TRU Discharged from UO_2 pins, kg	209	227	231	231	228	230
TRU Discharged from FFF pins, kg	285	402	435	454	458	457
TRU Discharged Total, kg	495	629	665	684	685	687
Net TRU generation, kg	-85.1	-42.4	-28.7	-14.5	-15.8	-16.0
Discharge Burnup, MWd/kg	68.3	59.0	57.1	56.7	56.5	56.3
Discharge Burnup, EFPD	1428	1233	1193	1184	1180	1177

Table 9. Poisoned CONFU Assembly: Materials Flow Summary (per 1 GWeY)

	Recycle Stage				
	1	2	3	4	5
Total HM Loaded, kg	17,996	19,486	20,539	20,916	21,019
Uranium Loaded, kg	17,334	18,770	19,784	20,147	20,247
TRU Loaded to FFF pins, kg	671	727	766	780	784
TRU Discharged from UO_2 pins, kg	218	232	239	241	242
TRU Discharged from FFF pins, kg	453	505	552	570	576
TRU Discharged Total, kg	671	737	791	811	817
Net TRU generation, kg	-0.8	9.7	24.8	30.8	33.0
Discharge Burnup, MWD/kg	61.3	56.6	53.7	52.7	52.5
Discharge Burnup, EFPD	1279	1231	1168	1147	1142

The results of the calculations presented above prove that a sustainable fuel cycle design is feasible. Table 8 and Table 9 show clearly that the TRU generation balance and the fuel cycle length are converging to constant values for both poisoned and un-poisoned CONFU assemblies. The TRU isotopic vector composition is getting stabilized (Table 10) as far as the most neutronically important isotopes are concerned. However, the buildup of some Cm (246,247,248,249) and Cf (249,250,251) isotopes does not saturate within the 5 recycles. The importance of these isotopes buildup lays in the fact that even their minute amounts can significantly complicate the fuel handling and reprocessing due to very high spontaneous fission (SF) neutron source.

1.4.1 Waste Characteristics

The potential environmental hazard characteristics of the fuel materials circulating in the sustainable fuel cycle based on the CONFU concept are summarized in Table 11 and Figures 23 – 26. All values in the Table 11 and in Figures 23 – 26 are normalized per cycle energy (in GWe Years) The CONFU fuel cycle environmental hazard characteristics were calculated at two time points between the recycles: at 5 years after discharge (fuel reprocessing) and at 7 years after discharge (new fuel fabrication). Only

the actinide contributions to the fuel characteristics were considered. As can be observed from Table 11 and Figures 23 – 26, the activity, decay heat load and radiotoxicity of the materials within the fuel cycle are also converging to equilibrium value. The SFS source is the only parameter that does not converge within the 5 cycles analyzed. As mentioned above, the SFS source can be one of the factors that limit the number of successive TRU recycles.

Table 10. Un-Poisoned CONFU Assembly: Actinides Isotopic Composition Vector (weight %)

Nuclide		Recycle Stage						
Z	A	0	1	2	3	4	5	6
92	234	0.00	0.51	0.98	1.29	1.45	1.49	1.44
92	235	0.00	0.04	0.21	0.38	0.51	0.58	0.60
92	236	0.00	0.02	0.06	0.12	0.20	0.27	0.34
92	238	0.32	0.14	0.12	0.11	0.11	0.11	0.11
93	236	0.00	0.00	0.00	0.00	0.00	0.00	0.00
93	237	6.64	5.43	4.45	3.87	3.56	3.37	3.29
93	239	0.00	0.00	0.00	0.00	0.00	0.00	0.00
94	238	2.75	8.04	9.92	9.99	9.33	8.55	7.86
94	239	48.65	25.83	22.03	20.95	20.26	19.82	19.31
94	240	22.98	26.58	23.33	20.84	19.34	18.52	17.86
94	241	6.93	10.61	9.31	8.30	7.67	7.29	7.01
94	242	5.03	11.65	16.49	19.90	22.40	24.08	25.55
95	241	4.65	5.69	5.25	4.62	4.17	3.92	3.70
95	243	0.02	2.83	3.61	4.15	4.53	4.76	4.99
95	242m	1.47	0.11	0.11	0.10	0.08	0.07	0.06
96	242	0.0000	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001
96	243	0.0050	0.0321	0.0392	0.0368	0.0327	0.0298	0.0279
96	244	0.4960	2.0244	3.1283	3.9429	4.5474	4.9208	5.2622
96	245	0.0380	0.3854	0.6748	0.8564	0.9679	1.0369	1.0691
96	246	0.0060	0.0874	0.2633	0.5050	0.7852	1.0657	1.3611
96	247	0.0000	0.0027	0.0123	0.0291	0.0513	0.0759	0.1025
96	248	0.0000	0.0003	0.0025	0.0085	0.0200	0.0366	0.0595
98	249	0.0000	0.0000	0.0001	0.0003	0.0008	0.0015	0.0024
98	250	0.0000	0.0000	0.0000	0.0001	0.0002	0.0004	0.0006
98	251	0.0000	0.0000	0.0000	0.0001	0.0002	0.0003	0.0006
Total:		100.00	100.00	100.00	100.00	100.00	100.00	100.00

Table 11. CONFU Assembly: Fuel Environmental Hazard Characteristics at the Time of Separation and Fabrication. (Normalized per 1 GWeYear)

		Recycle Stage				
		1	2	3	4	5
Separation Stage, 5 years after discharge	HM mass flow, kg	15018	17560	18197	18320	18436
	Radioactivity, Ci	7.73E+06	9.71E+06	9.97E+06	9.90E+06	9.86E+06
	Thermal Power, W	5.99E+04	1.04E+05	1.26E+05	1.37E+05	1.44E+05
	Ingestion Radiotoxicity, Sv	1.09E+10	1.75E+10	2.00E+10	2.08E+10	2.11E+10
	Total Neutron Source, #/sec	1.31E+11	3.45E+11	8.11E+11	1.77E+12	3.25E+12
Fabrication Stage, 7 years after discharge	TRU mass flow, kg	495	629	665	676	686
	Radioactivity, Ci	7.06E+06	8.89E+06	9.14E+06	9.08E+06	9.05E+06
	Thermal Power, W	5.61E+04	9.73E+04	1.18E+05	1.28E+05	1.34E+05
	Ingestion Radiotoxicity, Sv	1.03E+10	1.64E+10	1.88E+10	1.95E+10	1.98E+10
	Total Neutron Source, #/sec	1.20E+11	2.92E+11	6.02E+11	1.20E+12	2.10E+12

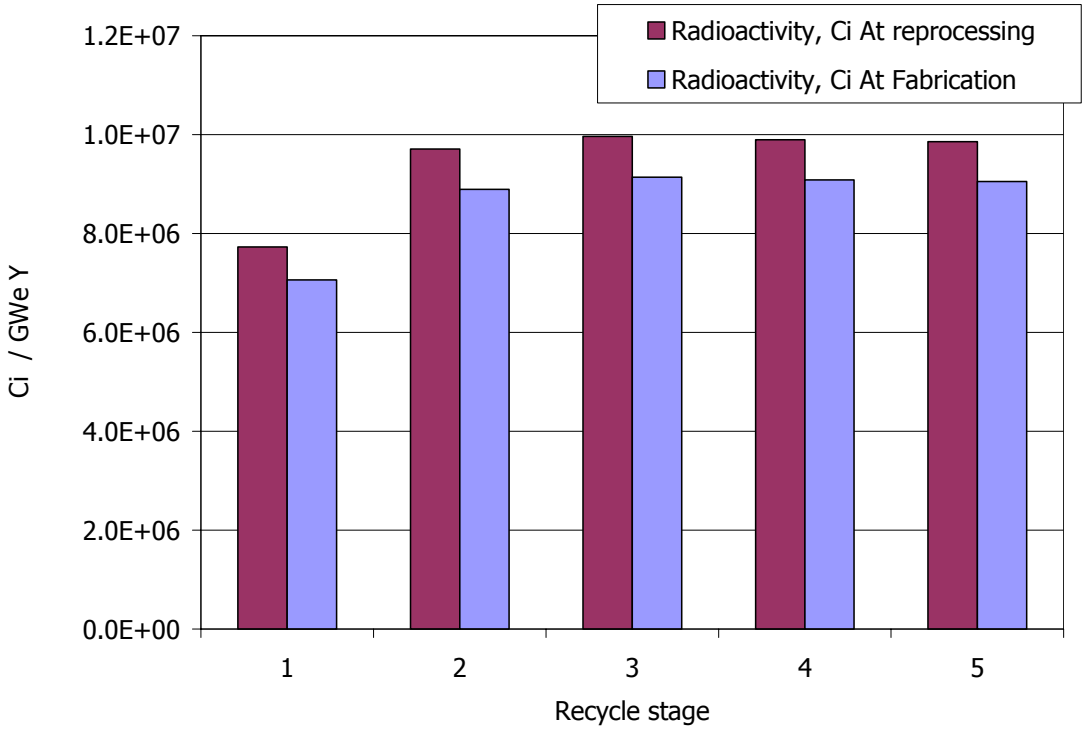


Figure 23. Radioactivity of the CONFU Fuel Assembly

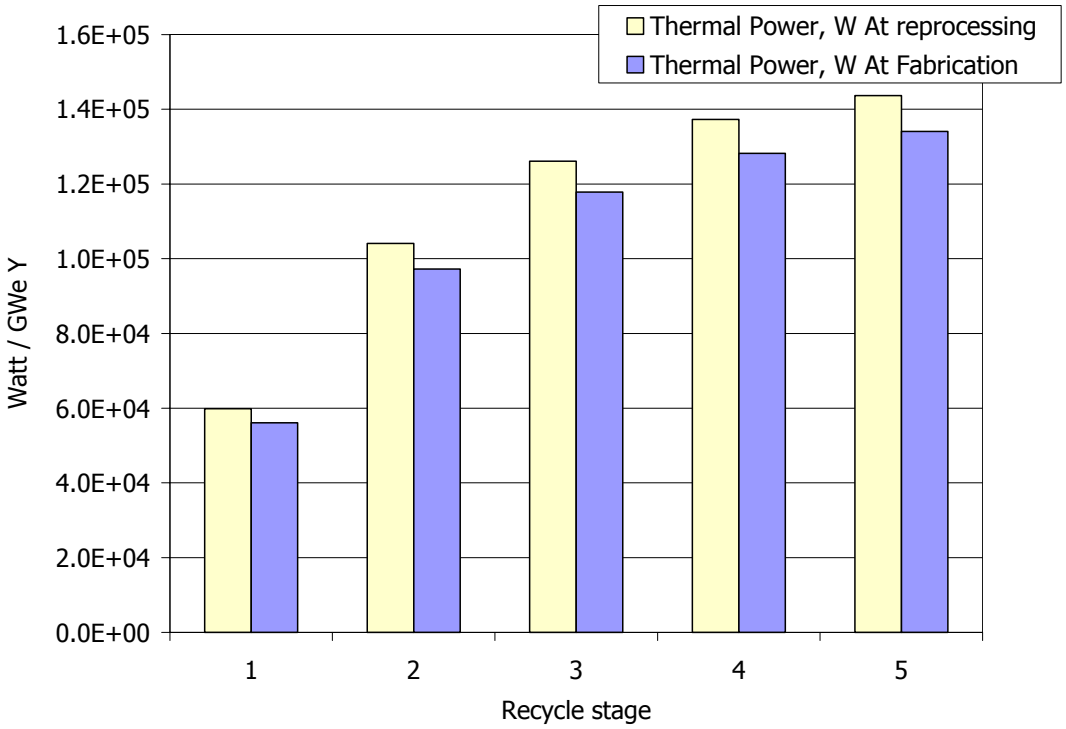


Figure 24. Decay Heat Generation of the CONFU Fuel Assembly

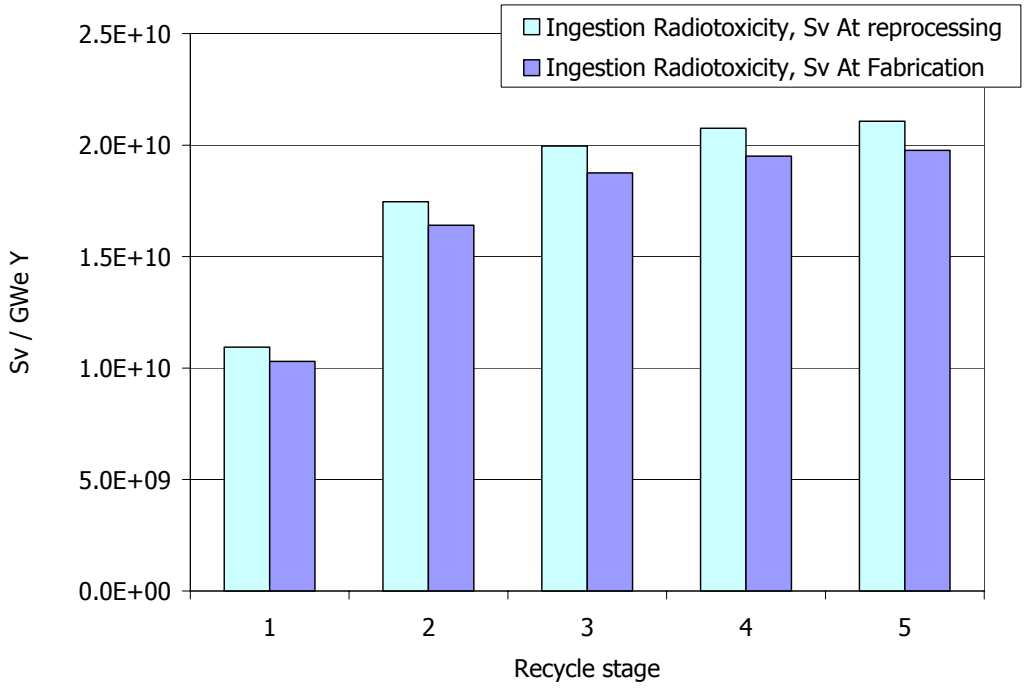


Figure 25. Ingestion Radiotoxicity of the CONFU Fuel Assembly

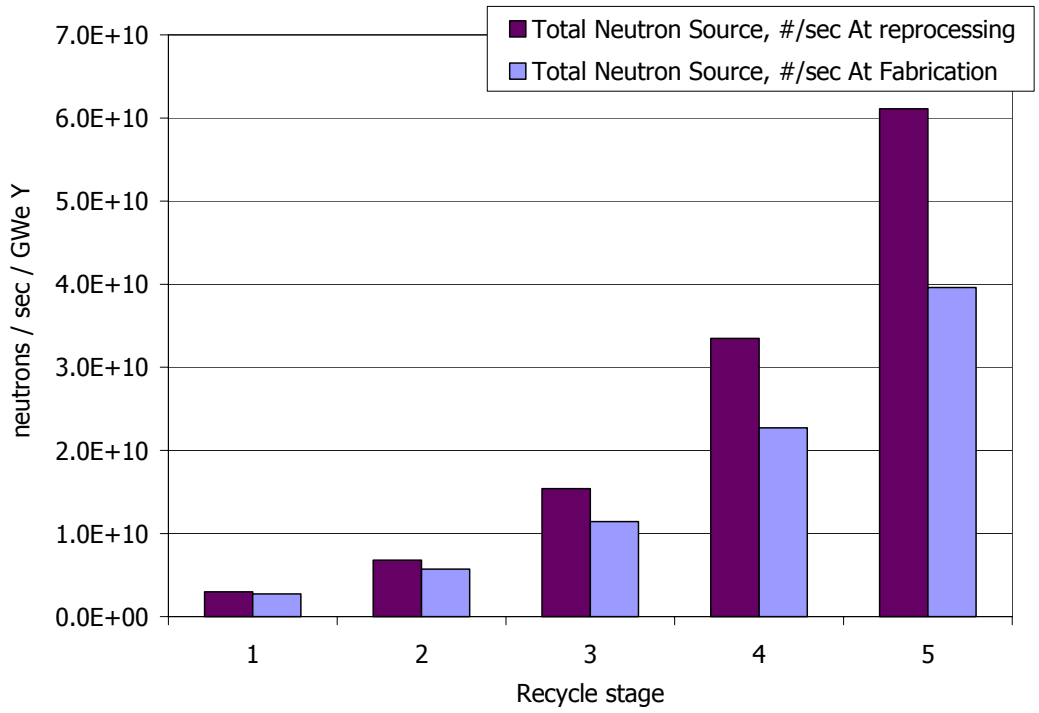


Figure 26. Total SF and (α,n) Neutron Source of the CONFU Fuel Assembly

The processes of TRU separation and fabrication are not perfect and have some limited efficiency. Therefore, complete TRU recycling is impossible because some TRU will inevitably leak to the environment as separation and fabrication processes waste streams. Additional calculations were performed to quantify the effect of TRU losses on repository load and compare it with conventional UO_2 spent fuel characteristics. The total TRU losses during each recycle stage were assumed to be 0.1%. We also assumed that all the losses had occurred at one time point of 5 years after the discharge. The waste stream characteristics were analyzed for the time interval between 0 and 1m years.

The results are reported in Figures 27 through 30. All the data summarized in Figures 27 – 30 are normalized per cycle energy in GWe Years. The results indicate that the activity, decay heat load and radiotoxicity of the waste streams from CONFU type fuel are up to 3 orders of magnitude lower than the same characteristics of the conventional once through UO_2 fuel cycle for the entire time interval considered.

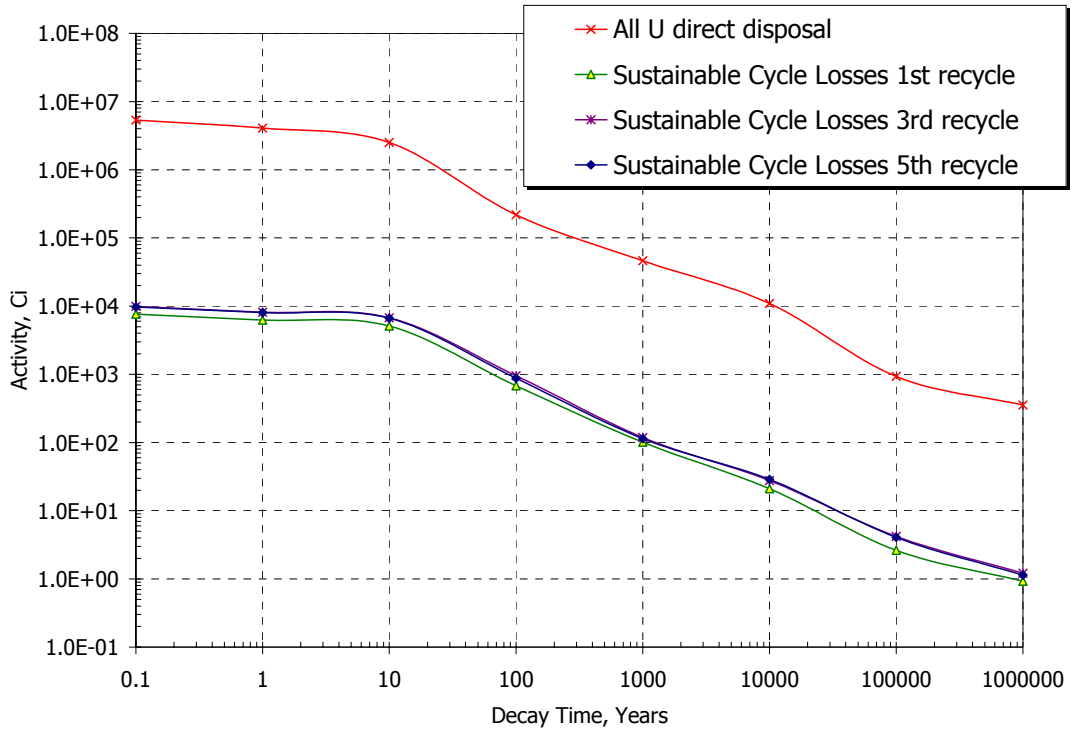


Figure 27. Losses from CONFU Assembly Recycling Process: Activity

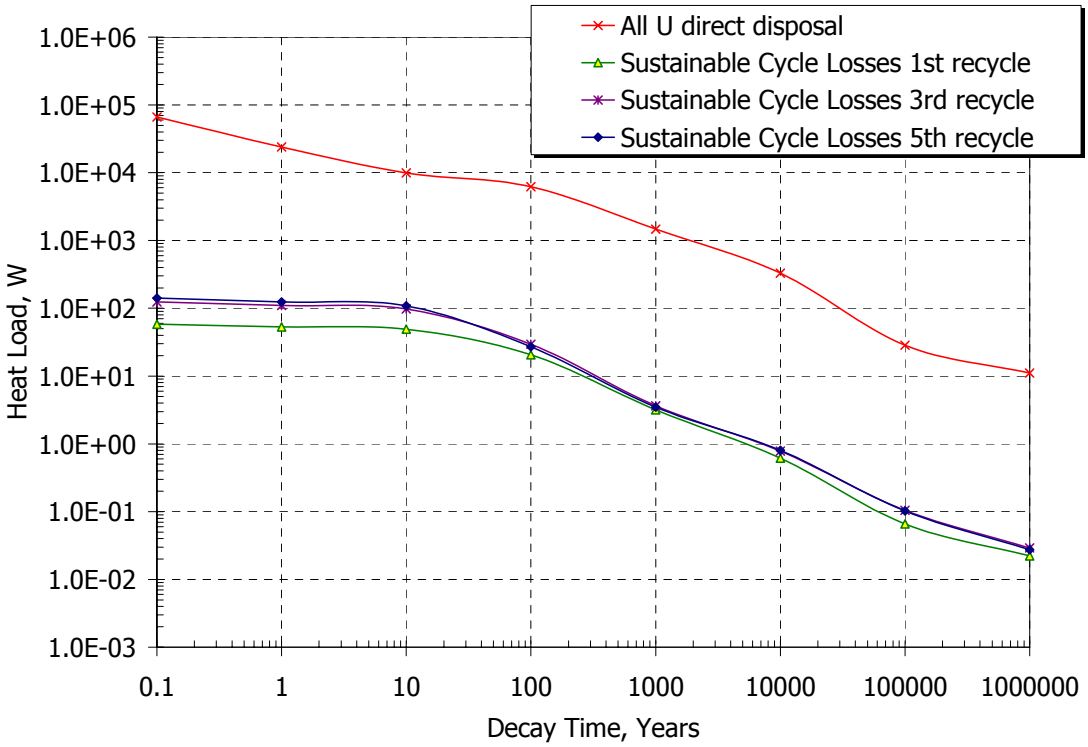


Figure 28. Losses from CONFU Assembly Recycling Process: Heat Load

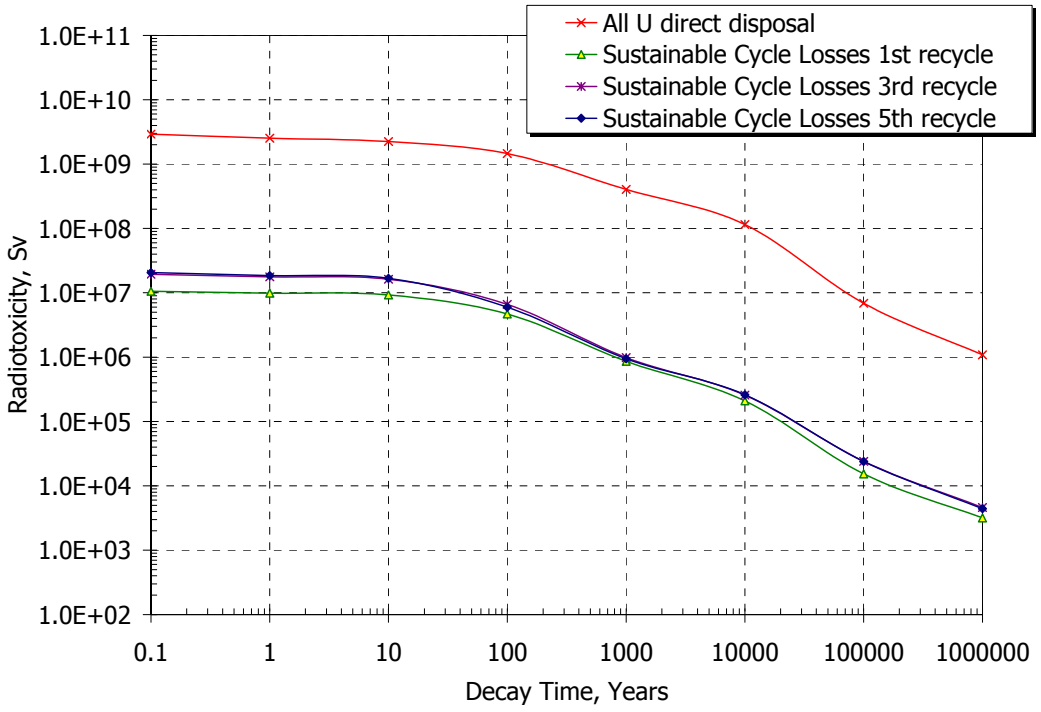


Figure 29. Losses from CONFU Assembly Recycling Process: Ingestion Radiotoxicity

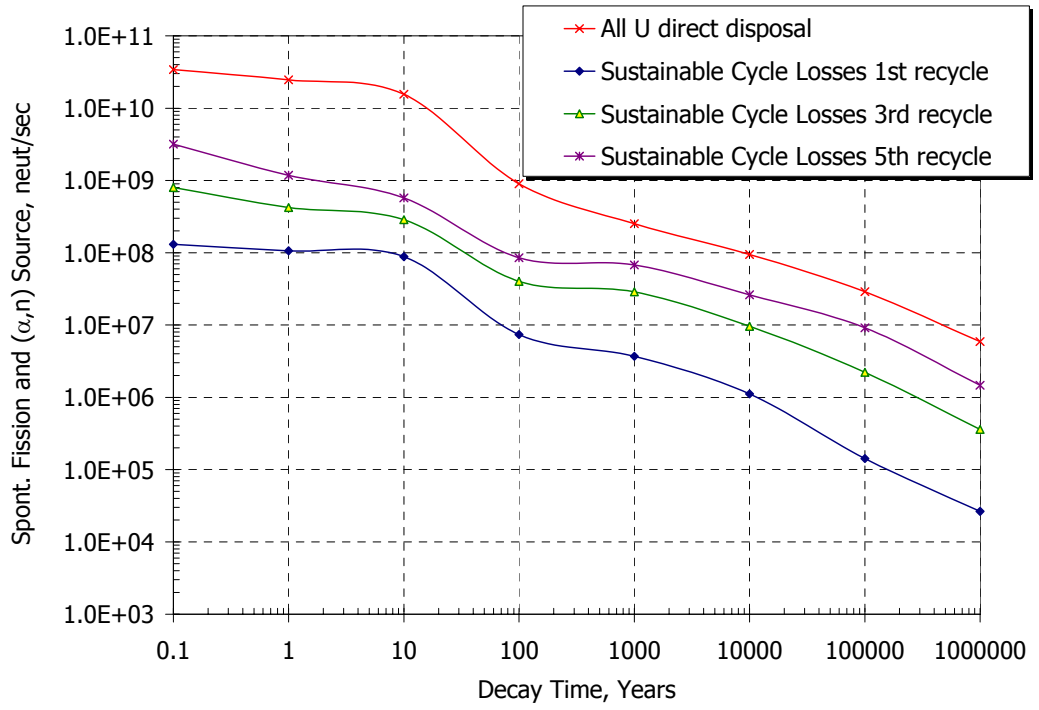


Figure 30. Losses from CONFU Assembly Recycling Process: Total SF and (α,n) Neutron Source

1.4.2 Reactivity and Control Characteristics

The CONFU assembly reactivity feedback coefficients were evaluated in order to assess the feasibility of retrofitting this concept into the current generation of PWRs. As in the previous parts of this study, the reactivity coefficients were calculated on an assembly basis at three time points – BOL, MOL and EOL with soluble boron concentrations of 1000ppm, 500ppm and 0ppm at BOL, MOL and EOL respectively. MOX and reference all-U assembly reactivity coefficients were also calculated on the same basis for comparison purposes.

Selected results of the CONFU assembly reactivity coefficients and soluble boron worth (BW) calculations are reported in Table 11. The results show that all the coefficients as well as BW fluctuate very slightly for different recycle stages. The DC tends to be more negative with increasing number of TRU recycles. The use of Er as a burnable poison somewhat improves the DC of the CONFU fuel. In general, all of the CONFU assembly reactivity coefficients and soluble BW differ only slightly from the reference all-U fuel, which indicates a good potential compatibility of the CONFU fuel concept with conventional PWR systems.

An evaluation of the effective delayed neutron fraction was performed in order to assess the effect of buildup of Cm isotopes with particularly small β_{eff} with the number of recycle stages. The results are shown in Figure 31. The CONFU assembly β_{eff} values at the beginning of fuel irradiation are moderately lower than corresponding values for the reference UO_2 assembly because of Pu239 fissions in the fertile free pins. This difference disappears with burnup.

The effect of small β_{eff} for Cm isotopes was not observed due to very small amounts of Cm in the fuel and its negligible contribution to total assembly power.

Table 12. CONFU Assembly Reactivity Coefficients Summary

				No Poison, Recycle Stages			With Poison, Recycle Stages		
		MOX	All-U	1	3	5	1	3	5
DC	BOL	-2.73	-2.03	-1.77	-1.90	-1.90	-1.88	-1.95	-1.98
	MOL	-2.87	-2.65	-2.01	-2.17	-2.19	-2.11	-2.19	-2.23
	EOL	-3.03	-3.08	-2.30	-2.41	-2.45	-2.33	-2.41	-2.44
MTC	BOL	-42.48	-11.26	-13.33	-15.26	-15.64	-17.01	-17.62	-18.06
	MOL	-54.36	-39.54	-32.96	-36.03	-36.80	-37.20	-38.46	-38.79
	EOL	-73.98	-66.29	-55.08	-57.40	-58.25	-58.46	-59.02	-59.66
VC	BOL	-112.4	-34.76	-38.59	-44.37	-45.34	-47.95	-49.92	-50.88
	MOL	-148.8	-119.0	-96.35	-106.0	-107.9	-107.4	-111.5	-113.3
	EOL	-204.0	-205.9	-161.2	-172.9	-175.5	-172.1	-176.9	-178.9
BW	BOL	-2.70	-6.11	-5.16	-5.51	-5.54	-5.41	-5.64	-5.70
	MOL	-3.17	-7.09	-5.61	-6.02	-6.07	-5.81	-6.08	-6.13
	EOL	-3.91	-9.51	-6.98	-7.45	-7.57	-7.10	-7.43	-7.53

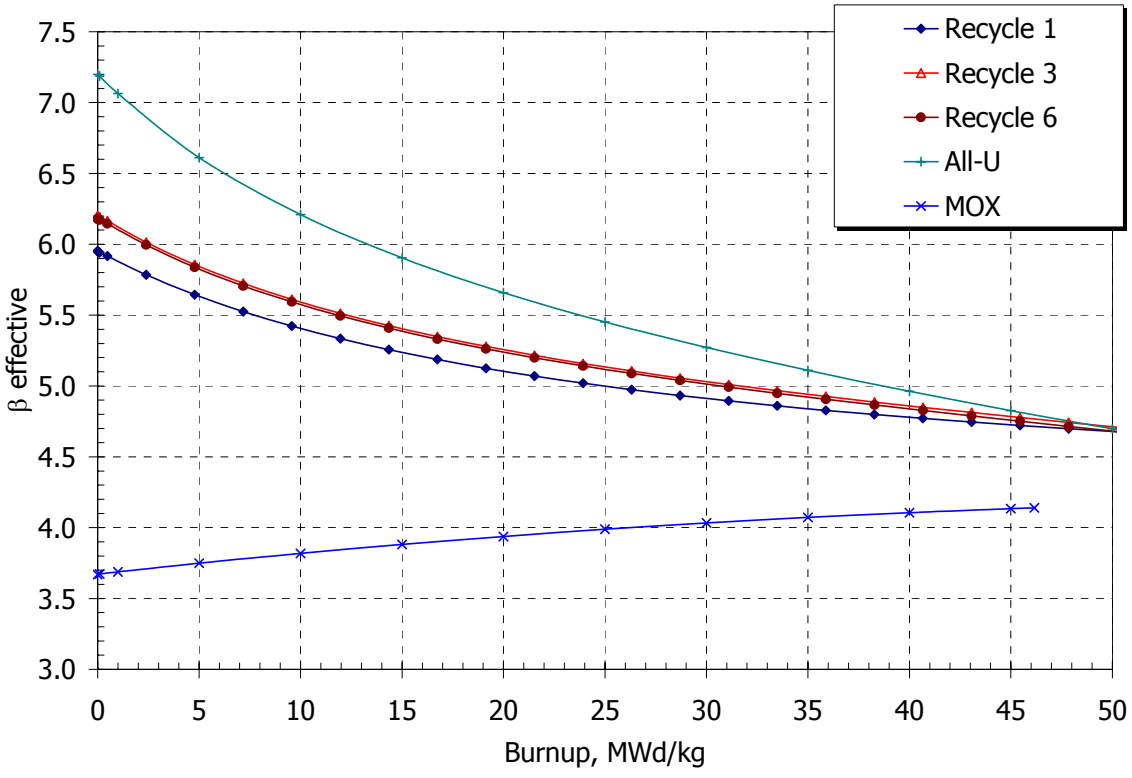


Figure 31. Effective Delayed Neutron Fraction × 10^3 vs. Burnup for CONFU Assembly

1.5 Summary and Conclusions

In this work, we evaluated the potential of fertile free fuel concepts to reduce the stockpile of TRUs in the spent fuel of LWRs and to design a sustainable fuel cycle for conventional PWRs with complete TRU recycling.

The benchmark calculations performed indicate that the computational tools used are capable of producing results with the accuracy sufficient for the purposes of the study.

Homogeneous and heterogeneous fuel assembly configurations were evaluated and compared in terms of TRU destruction efficiencies, reactivity feedback coefficients and spent fuel characteristics. The heterogeneous core concept implying two different assembly types with different relative amounts of Pu and MA cannot improve the efficiency of TRU destruction beyond that of the homogeneous core with only one type of the assemblies.

The once-through TRU burner based on the FFF can destroy over 50% of initial TRU per path. An additional stage of TRU recycling is less useful because of the much lower destruction efficiency (less than 35%).

The spent fuel characteristics are almost identical for homogeneous and heterogeneous core approaches. Substantial reduction of the current LWR spent fuel radiotoxicity and decay heat is impossible using one stage recycling and burning in the FFF type fuel assemblies. The energy normalized spent fuel characteristics for the once through TRU burner are at most breakeven with all-U fuel values.

The reactivity coefficients of the FFF based TRU burner have negative values although further evaluation is required to ensure feasibility of the core with 100% FFF assembly loading. The uncertainty is associated with the difficulty to evaluate a correct soluble boron concentration. The soluble BW is up to 3 times lower for the FFF assembly in comparison with typical PWR values. As a result, a redesign of the reactor control

features is likely to be required, for example, use of enriched Gd or/and Boron as a reactivity control materials or increased number of control rods.

In addition, the feasibility of PWR core with 100% TRU loading in the FFF assemblies is uncertain due to considerably lower effective delayed neutron fraction in comparison with typical UO_2 and MOX fuel.

The evaluation of the sustainable cycle with zero net generation of TRU was based on the Combined Non-Fertile and Uranium (CONFU) assembly concept where some of the uranium fuel pins are replaced with FFF pins destined for destruction of TRU generated in the previous cycle.

The results indicate that the CONFU type of assembly can be designed to achieve an equilibrium state in terms of net generation of TRU and at the same time have acceptable reactivity control and thermal-hydraulic characteristics.

The impact on the environment of the CONFU based fuel cycle with complete recycling of TRU is limited by the materials losses during reprocessing. The CONFU based fuel cycle waste stream have up to 3 orders of magnitude lower values for the radiotoxicity and decay heat than conventional once-through All-U fuel cycle waste.

The buildup of some Cm and Cf isotopes that do not saturate in LWR neutron spectra imposes potential limits on the number of TRU recycling stages due to a large spontaneous fission neutron source which can significantly complicate spent fuel handling, separation and fabrication procedures.

The effect of Cm buildup on the effective delayed neutron fraction is negligible. The β_{eff} values for the CONFU assembly are comparable with the reference PWR UO_2 fuel.

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APPENDIX A

A Survey of Inert Matrix Materials as Hosts of Actinide Nuclear Fuels

Currently there are increasing interests in using inert matrix fuel (IMF) to reduce, if not eliminate, the accumulation of plutonium and minor actinides worldwide. The use of inert matrix materials poses design challenges because the absence of uranium in the fuel significantly influences the burnup reactivity swing, the reactivity coefficients and the kinetic parameters of the fuel.

The chosen IMF material should be neutron transparent, have high chemical compatibility with the cladding and the coolant, have good resistance to irradiation damage, have good heat capacity, high melting or phase transformation temperature, low thermal expansion, good thermal conductivity, and good mechanical properties and be economically reasonable. Fabrication and availability of starting materials are important considerations. Furthermore, the new fuel should be able to maximize the plutonium burnup and, if applied to LWR, be easily incorporated into the present LWR fuel cycle.

For the elements meeting the basic neutronic requirements for use as inert matrix materials, several formats are possible (among metallic, silicide, nitride, carbide and oxide), and several crystalline structures are desired (among peroskovites, fluorites, yag, and rutile).

The metal, carbide and nitride fuels are not readily acceptable for LWRs because of their chemical interaction with water. An oxide is preferred because of the plentiful fabrication and operating experience. We also choose the fluorite as the crystalline structure due to the large experience base. Another reason is its ability to incorporate actinides, rare earths and fission products.

The candidate inert materials surveyed are zircon $ZrSiO_4$, stabilized zirconia $(Zr,Y)O_{2-x}$ and $(Zr,Ca)O_{2-x}$, Al_2O_3, MgO, spinel $MgAl_2O_4$, CeO_2, monazite $CePO_4$, SiC and some metals.

Some of the candidates could be rejected: zircon ($ZrSiO_4$) because of its dissociation upon annealing to high temperatures and its extensive swelling under radiation; Al_2O_3 because of amorphization and large swelling; MgO because of its disintegration in the event of cladding failure under PWR conditions; monazite ($CePO_4$) because of poor radiation stability due to fission product impact and poor thermal conductivity. There are difficulties with CeO_2 because of its polygonizatin and temperature dependent swelling. It seems that stabilized zirconia is a good choice, with CeO_2 as a backup.

Zirconia can be stabilized by either calcium or yttrium oxide. Y_2O_3 has negligible neutron induced swelling. Yttrium stabilized zirconia (YSZ) has high chemical stability, high melting temperature, high irradiation stability. It is an excellent actinides host matrix. CaO is also a good candidate to stabilize zirconia. It has very good thermo-mechanical properties.

The major drawback of a zirconia matrix is its low conductivity, but spinel can be added to compensate for this. Spinel exhibits partial amorphization and polygonization and is unstable under fission product impact. We can use a hybrid fuel concept to localize the irradiation damage. Plutonium is incorporated with stabilized zirconium to form a fluorite phase, which is subsequently dispersed into a spinel matrix. Fission fragments can be localized by YSZ in the fissile phase.

The fuel can be in the format of a CERCER, a CERMET, or a Solid Solution Pellet (SSP). SSP is the result of homogeneous mixing of fissile isotopes into an inert matrix, the resultant IMF is a single-phase material. For effective consumption of Pu, very high burnup of initial inventory (>40%) is desired. The traditional solid solution of ceramics or alloy fuel is not appropriate for this burnup level. Hybrid fuel concept like CERCER or CERMET is necessary.

The multi-phase CERCER and CERMET have several advantages in providing better performance. By using two-phase plutonium materials the integrity of the inert matrix is better preserved during irradiation. Dispersed fuel is preferred to solid solution because it can localize the damage to matrix phase thus can avoid degradation of its thermal conductivity, which results in higher achievable burnup. The drawbacks are: more complicated manufacturing and potential hot spots and absence of thermodynamic stabilization.

Although no common strategy for uranium-free fuels has emerged, the fuel concepts for CERCER fuel seem to be converging to $MgAl_2O_4$ (spinel) as the matrix, with plutonium dissolved in either CaO or Y_2O_3 stabilized zirconia as fuel. In some effort cerium oxide is used instead of zirconium oxide, and $Y_3Al_5O_{12}$ is used instead of spinel ($MgAl_2O_4$) as inert matrix materials, but the dominant trend is to use zirconium oxide and spinel.

The additives of ThO_2 and UO_2 effectively increase the negative Doppler coefficient of the inert matrix fuel. With these additives, however, the plutonium transmutation rate decreases, because ^{232}Th and ^{238}U produce the fissile nuclides ^{233}U and ^{239}Pu, respectively. Core burnup calculations show that the fuel ROX cores can transmute much larger amounts of plutonium annually, i.e. 1.5 to 2.0 times as much as the MOX core and that a fully ROX loaded one GW electrical power class LWR is capable of transmuting nearly one ton of Pu every year.

In summary, for LWR applications, it is found that ceramic oxides are more promising than other materials, although there is still no consensus in the literature as to the best host material. Zirconia (stabilized by yttria or calcia) and cerium oxide are considered most promising as hosts for the actinide oxides. For zirconia fuels, another material with a more favorable thermal conductivity, such as spinel ($MgAl_2O_4$), would be needed in the mixture to allow the fuel to operate at acceptably low temperatures. The sensitivity of spinel to fission product damage necessitates the use of a dispersion of actinide-host particles of a relatively large size, 150-200 μm, in a continuous matrix of spinel.

The considerations presented in this summary will be elaborated in a forthcoming report (Ref. 5).

APPENDIX B

Double-Heterogeneous Effect Evaluation and Codes Benchmarking:

Evolution of Selected Actinides Number Densities with Burnup

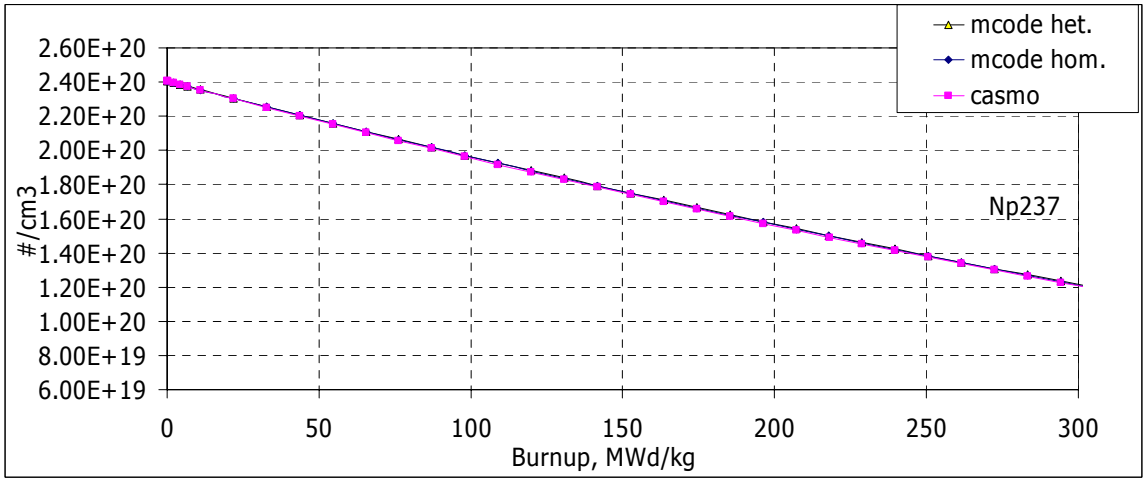

Figure 1. Np237 Number Density vs. Burnup

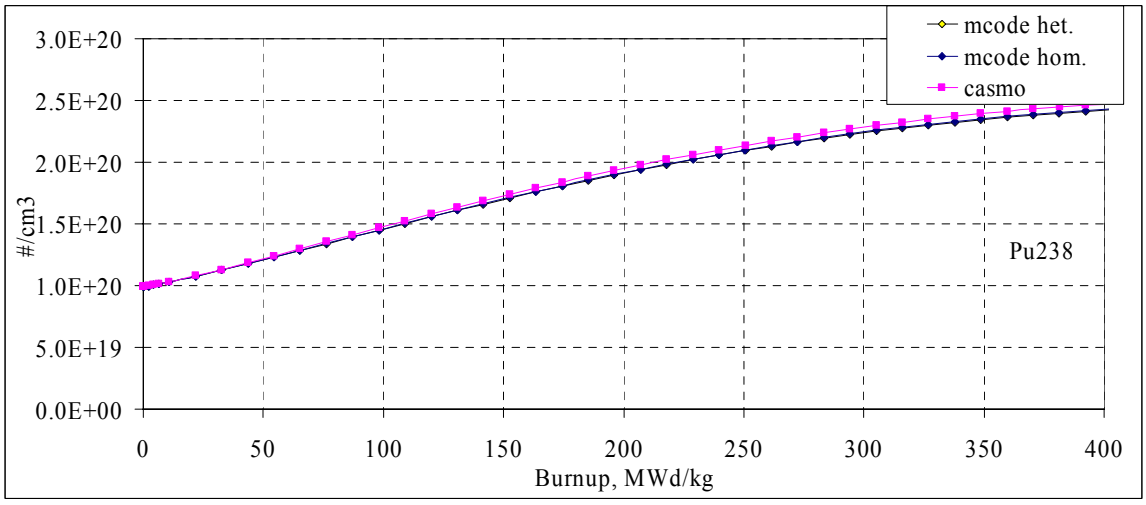

Figure 2. Pu238 Number Density vs. Burnup

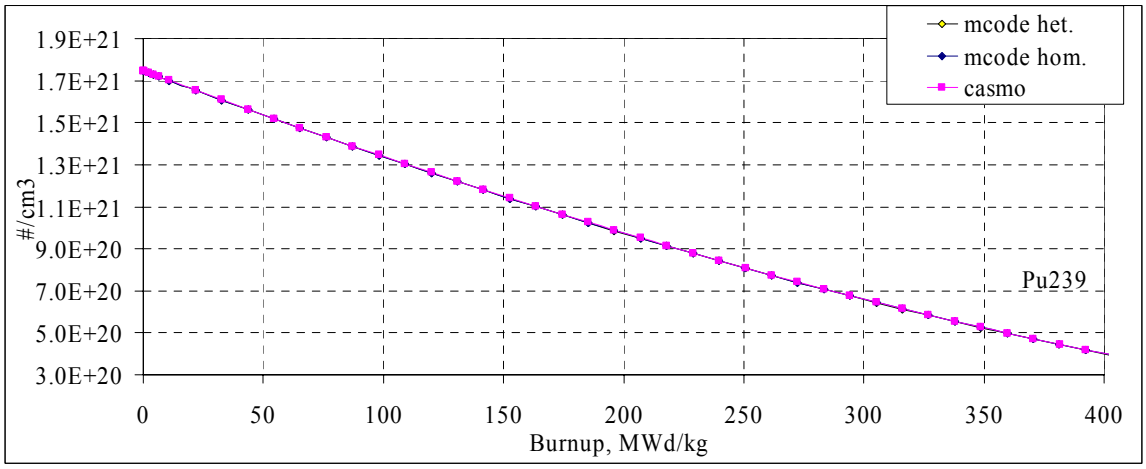

Figure 3. Pu239 Number Density vs. Burnup

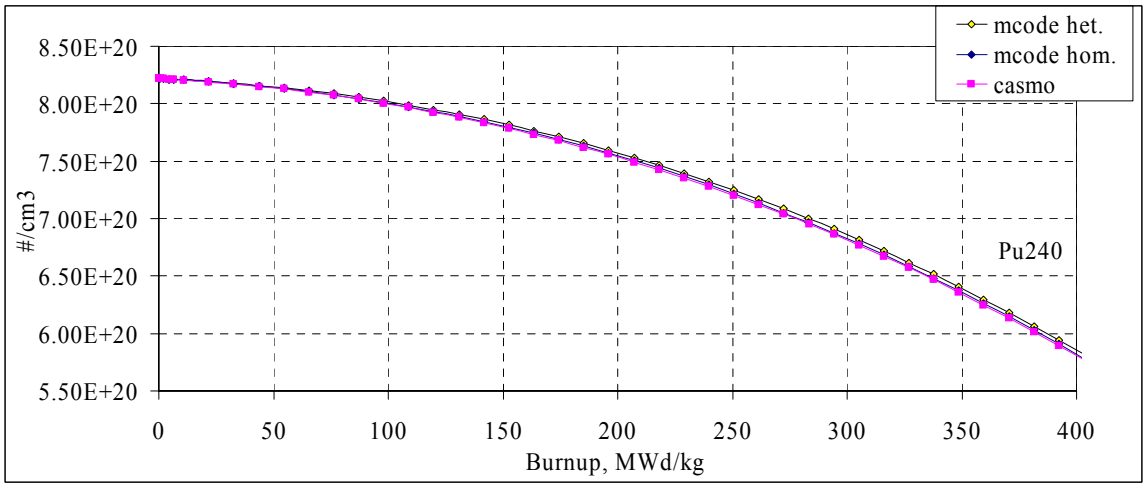

Figure 4. Pu240 Number Density vs. Burnup

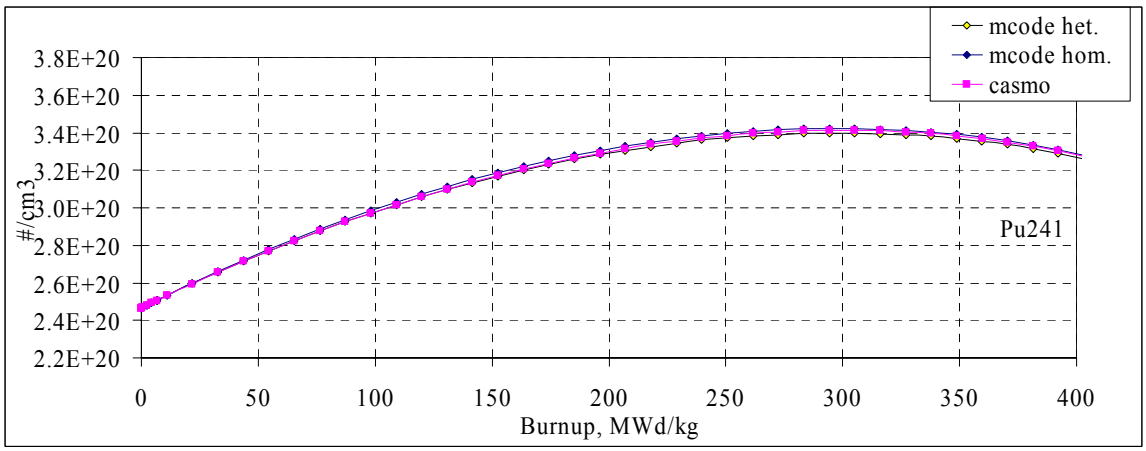

Figure 5. Pu241 Number Density vs. Burnup

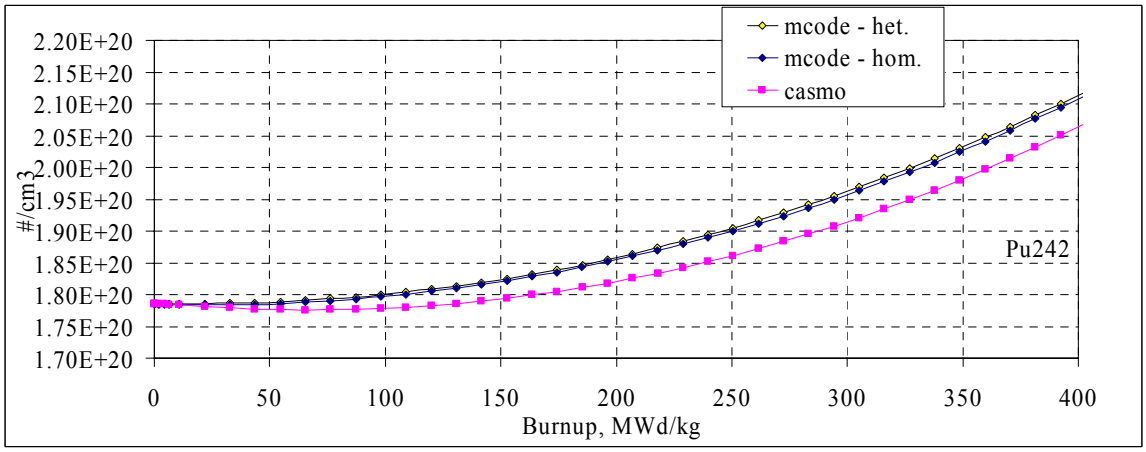

Figure 6. Pu242 Number Density vs. Burnup

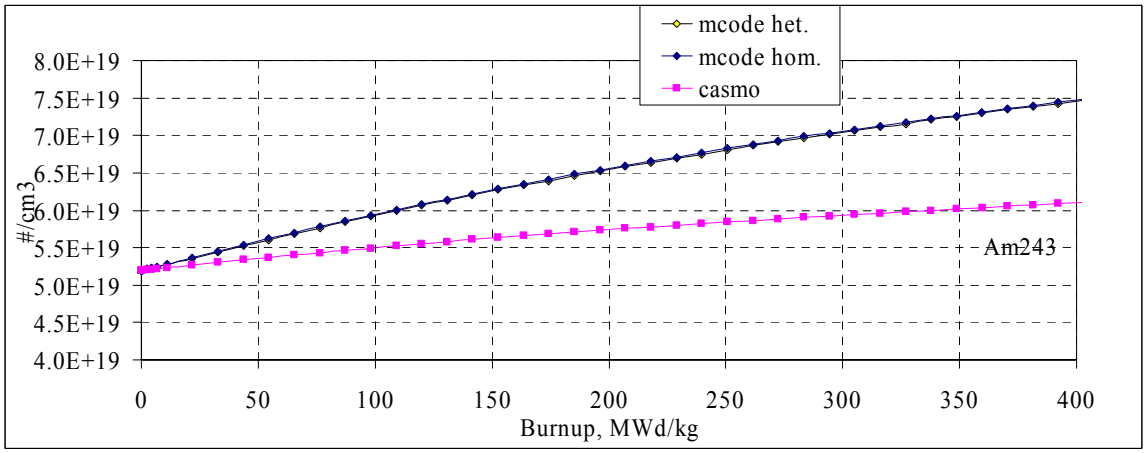

Figure 7. Am243 Number Density vs. Burnup

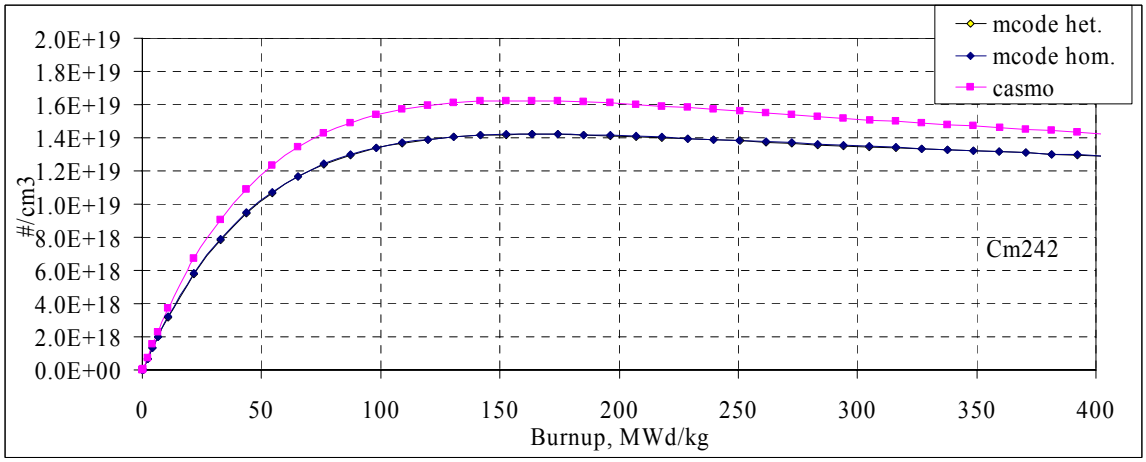

Figure 8. Cm242 Number Density vs. Burnup

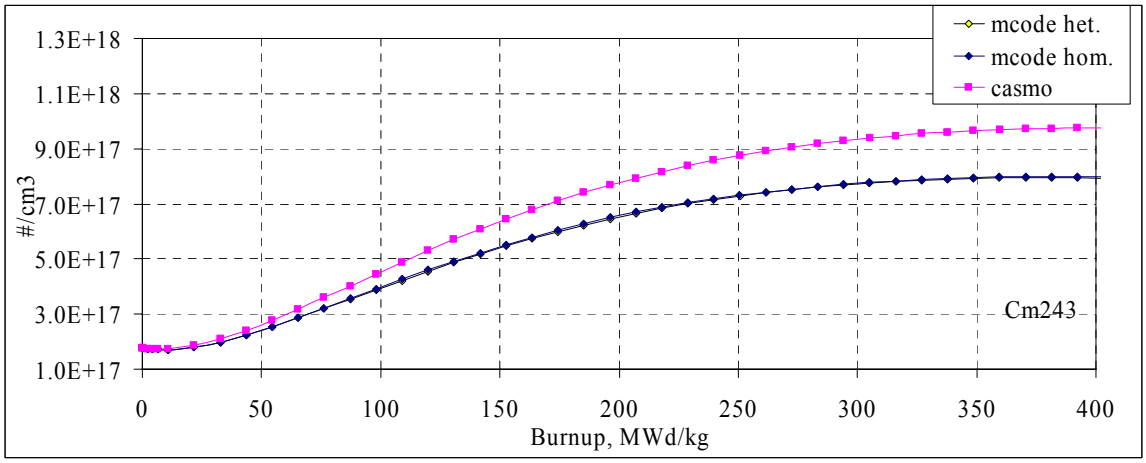

Figure 9. Cm243 Number Density vs. Burnup

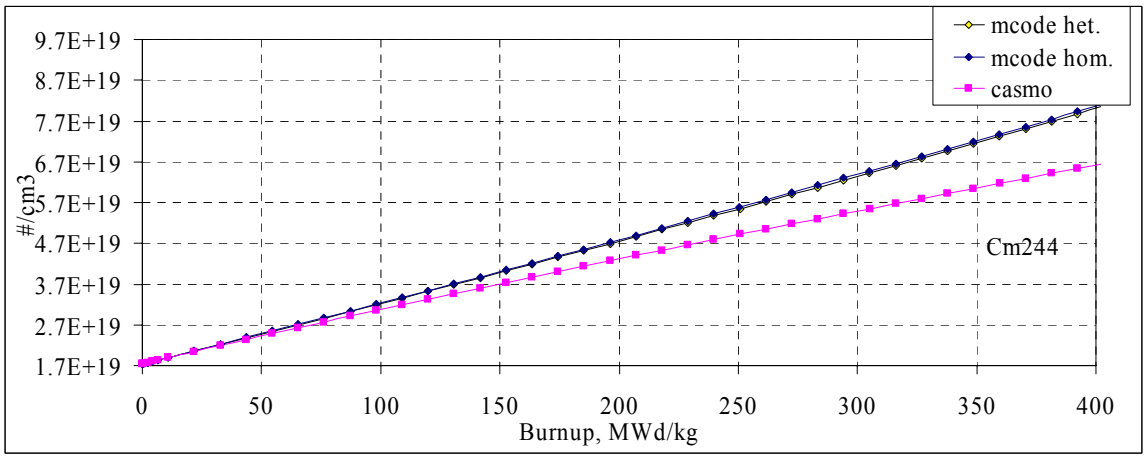

Figure 10. Cm244 Number Density vs. Burnup

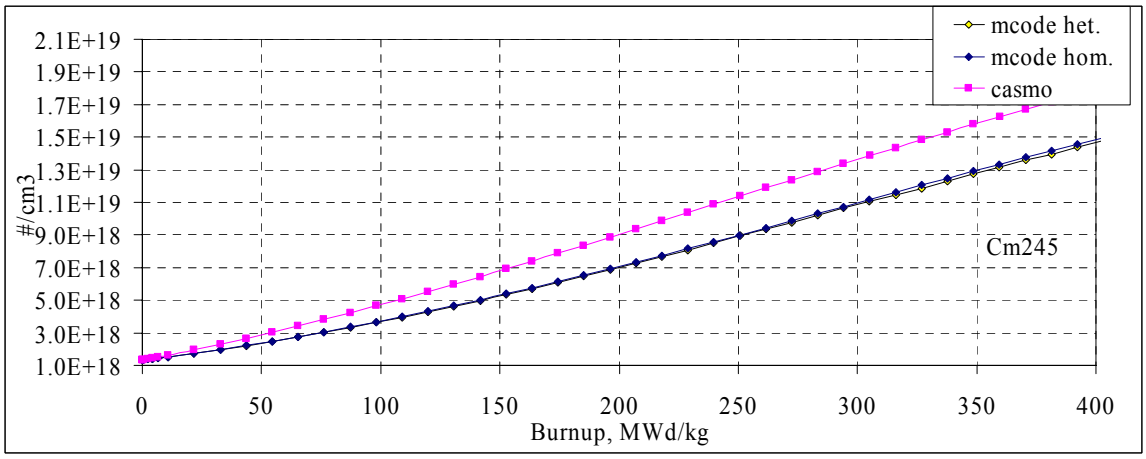

Figure 11. Cm245 Number Density vs. Burnup

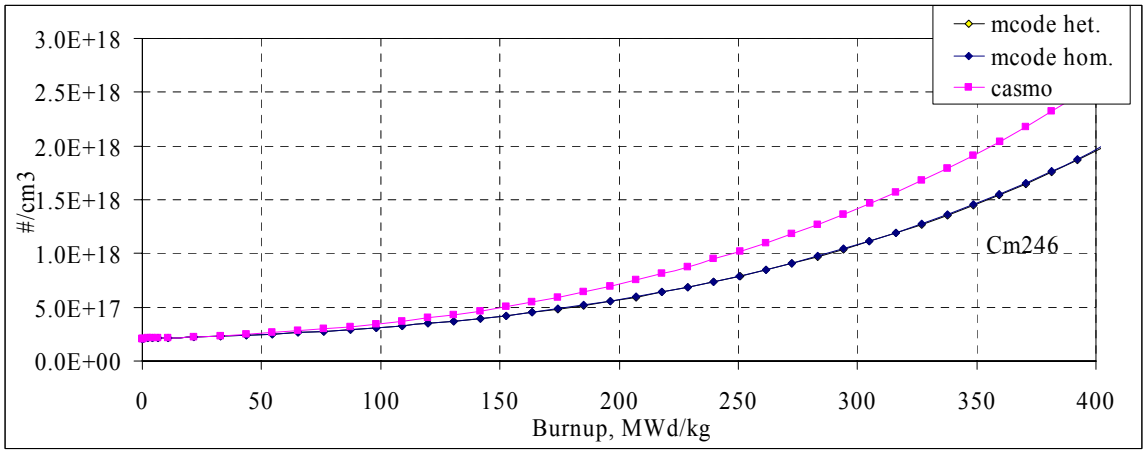

Figure 12. Cm246 Number Density vs. Burnup